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The influence of the unsaturated zone on the upward transport of radionuclides in soils

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Summary

The transport of radionuclides from the deep soil to the surface soil is an important part of biosphere modelling. It is in the root zone of the surface soil that radionuclides are accessible for plants and thereby can be taken up in the food chains, leading to a dose to man. The upwards transport through the soil is of importance for assessing the radiological consequences of underground waste disposal where transport in the groundwater is the primary source term for radionuclides coming to the biosphere.

In this study the effect of transient hydrological conditions on the upward transport of radionuclides through soils has been studied. The effect of varying soil properties, climate conditions have been considered as well as the effect of a fluctuating groundwater level. Theses studies have been performed with advanced mechanistic models. To investigate the influence of model complexity, the results have also been compared with the results obtained with a more simple compartment model.

It was shown that the soil characteristics influences the radionuclide concentration; an increased hydraulic conductivity leads to increase in the concentration in the root zone. The climatic conditions were shown to be of major importance, a drier climate results in increased radionuclide concentration in the root zone.

A dispersion dependent on both velocity and saturation leads to a more effective upward transport of radionuclides to the root zone than if dispersion is assumed to be dependent only on the saturation.

The boundary condition used in the case with varying groundwater level may be more realistic than the boundary condition applied for the case with a constant groundwater level. All calculations with varying groundwater level gave lower radionuclide concentration in the root zone.

Sorption is redox sensitive for many radionuclides and the redox potential in the soil will be affected by the degree of water saturation. The performed calculations did, however, not result in any significant change in the radionuclide concentration in the root zone due to variation in saturation.

A comparison between the results of the two models show that the compartment model in all studied cases predicts a higher annual average radionuclide concentration in the root zone than the numerical model. Annual variations in soil water flow were not included in the compartment model. During the summer the concentration in the root zone may be several times higher than the annual average. This may be important for plant uptake, since this increased concentrations coincides with the plant growing season. The calculations made with the simple compartment model also show that these types of models have an inherent dispersion that depends on the size of the compartments. Various methods to compensate for this inherent dispersion have been investigated, but additional work remains.

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1 Introduction

1.1 Background

The transport of radionuclides from the deep soil to the surface soil is an important part of biosphere modelling. It is in the root zone of the surface soil that radionuclides are accessible for plants and thereby can be taken up in the food-chains, leading to a dose to man. The upwards transport through the soil is of importance for assessing the radiological consequences of underground waste disposal where transport with the groundwater is the primary source term for radionuclides coming to the biosphere.

The major transport mechanism in soils is generally assumed to be the transport of dissolved radionuclides with flowing water - advection. Thus the movement of the water in the soil is of great importance. However, the movement of soil water is a complicated process, especially in the unsaturated zone, where it is affected by precipitation, evapotranspiration, capillary rise and groundwater level fluctuations. The radionuclide transport is also influenced by dispersive processes, e.g. due to a variation in flow between different parts of the soil and molecular diffusion.

In biosphere modelling compartment models are often used to describe the transport of radionuclides in soils. In these models the soil at different depth is described by two or more compartments, in which an instantaneous mixing is assumed. This gives a low degree of spatial resolution, which will have an important effect on the description of the dispersive processes. The transfer between the compartments is described by simple relationships taking into consideration average water flow rates, sorption, etc. These relationships are usually based on the temporal averaging of the soil hydrology, e.g. no consideration is taken to the varying degree of saturation over the year and that the upward and downward flows may occur at different degree of saturation. The saturation influences a number of important transport processes, e.g. diffusivity, dispersion and for some nuclides also sorption due to the effect of saturation on the redox conditions.

1.2 Purpose and scope

In a previous study [*Elert et al., 1990*] it was found that the hydrological conditions in the unsaturated zone was of importance for the upward transport of radionuclides. It was shown that the upward flow of water was the primary processes for transport of radionuclides to the root zone, but dispersive processes may also be effective for the upward transport of radionuclides. In the previous study the groundwater level was assumed to remain at a constant depth.

The purpose of project is to study the effect of transient hydrological conditions in the soil on the upward transport of radionuclides through soils considering a varying groundwater level. Also the effect of the varying hydrology on the chemical conditions, which in turn may influence the radionuclide sorption, was studied. These studies have been performed with advanced mechanistic models. The results have been compared with the result obtained by more simple models to investigate the influence of model complexity.

In this study the hydrological conditions in the soil in the form of time-varying water saturation levels and water flow rates have been calculated using a hydrology model. The input data of the model does not represent a specific site, but are taken from meteorological data and soil property data characteristic for Sweden. Two different soil types and two different climatic conditions have been studied. Furthermore, a set of hydrological calculations have been performed using a seasonally varying groundwater level.

The results of the hydrology calculations have been used as input to a radionuclide transport model. The transport of both non-sorbing and slightly sorbing radionuclides have been investigated. Variations in saturation will also lead to a variation in redoxpotential. This may influence the mobility of many radionuclides. One aim of the report was to investigate whether a varying redox potential in connection with a varying groundwater level may give rise to "pump-effects", i.e. if a higher mobility during the parts of the year with an upward water flow than during the parts of the year with a downward water flow, can give rise to significantly increased concentrations in the root zone.

The biosphere transport of radionuclides must often be considered over very long periods of time in safety assessments of radioactive waste repositories. In this case it is very difficult or even impossible to use detailed models, furthermore the necessary data for using such models may not be available. Instead compartment models using annual average values are used. Since the processes transporting radionuclides in soils are very complex and are often non-linear. Thus, it is not evident that the use of arithmetic averages in a simplified model will give correct results. Instead some form of weighted averages may need to be used. A purpose of this study has been to examine methods to derive such weighted or effective averages.

2 Modelling of hydrology

2.1 Model setup

In order to describe radionuclide transport in soil, information is needed on the water flow and saturation and how it varies in time and space. Experimental data could be used, but all necessary data are rarely available, for example water saturation degree and water flow as a function of time at different depths of the soil. Instead we have chosen to derive these values by hydrological modelling. This will give data with a spatial and temporal discretization suitable for the radionuclide transport calculations.

The computer code TRUST [*Narasimhan, 1975*] was used for the hydrological modelling. This code solves the saturated and unsaturated flow equations using the integrated finite difference method. The basic input data needed to describe the soil characteristics are the porosity, the capillary potential curve and the hydraulic conductivity as a function of saturation. Also, data describing infiltration, precipitation and the geometry of the studied soil column are needed. Furthermore, boundary conditions at the top and at the bottom have to be provided in the form of water flow or pressure.

2.2 Input data

The hydrology of a soil column has been model assuming a simple geometry. Two cases have been defined, one with a groundwater level at a constant depth of 1 meter and one with a groundwater level varying between depth of 0.7 meter and 1.3 meter. In the latter case the modelled domain is a 1.3 m deep soil column with a 0.6 m zone in the bottom where the groundwater level is fluctuating during the year, see Figure 2.1. In both cases the root zone is assumed to comprise the upper 0.3 meters.

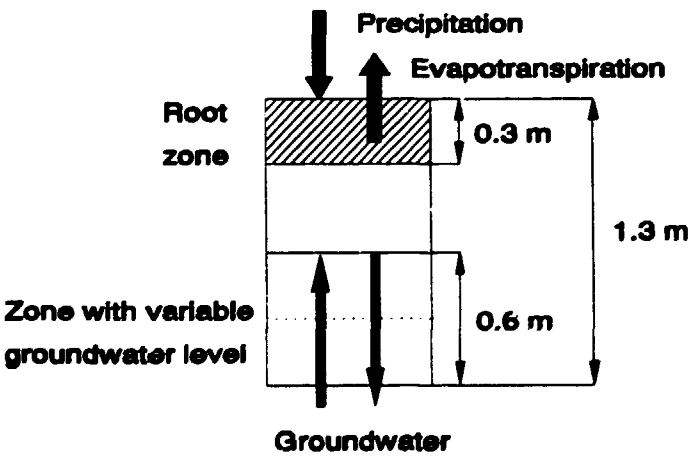

Figure 2.1 The geometry of the studied case.

The soil moisture characteristics or capillary potential curve was taken from experimental values for Swedish soils [*Andersson and Wiklert, 1972*] and represent an average for soils with a clay content of 3-5%. Since no measured values of the saturated hydraulic conductivity were provided for the chosen sample, we have estimated it to $1 \cdot 10^{-6}$ m/s, based on the particle size distribution. As a variation a higher saturated hydraulic conductivity, 10^{-5} m/s, was used in some calculations. The hydraulic conductivity of a soil is a function of the water saturation. In unsaturated porous media water flow occurs only as long as continuous pathways for water exists. Since the largest pores, which have the lowest flow resistance, will be emptied first, the hydraulic conductivity will decrease rapidly with decreasing saturation. The hydraulic conductivity as a function of saturation degree has been calculated from the experimentally determined capillary potential curve using the method proposed by *Green and Corey, [1971]*. The data used in the calculations are shown in Figures 2.2 and 2.3.

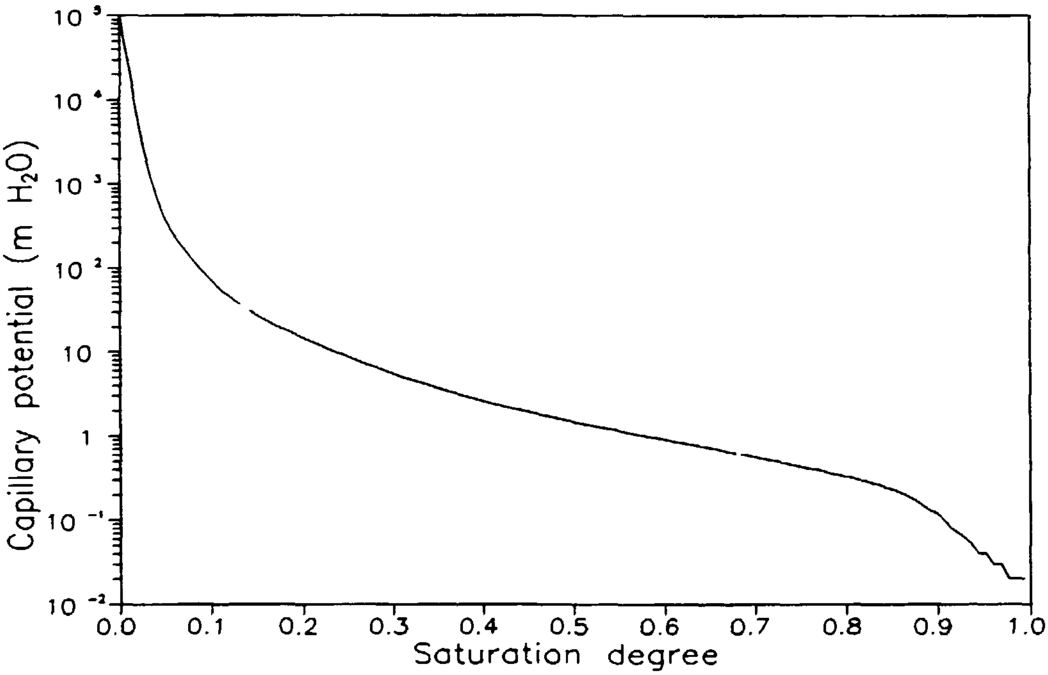

Figure 2.2 *The capillary potential as a function of saturation degree.*

The boundary condition at the top is provided by the infiltration and evapotranspiration data varying on a monthly basis. The precipitation data are taken from the weather stations in Ställdalen and Malmslätt. In most parts of Sweden the ground is frozen at least some parts of the winter. This will affect the water transport in two different ways. Firstly, the frozen soil water will not be mobile and secondly, a substantial amount of the precipitation will not infiltrate but produce surface run-off at the time of snow melt. In order to simplify the calculations no frost in the ground is assumed and furthermore no accumulation of snow.

The evapotranspiration is based on data from *Eriksson [1981]*, and is assumed to be evenly distributed throughout the 0.3 meter thick root zone. In order to avoid excessive drying of the soil, the evapotranspiration rate is reduced below a capillary water potential of -50 meters and is assumed to stop below the wilting point, -150 meters.

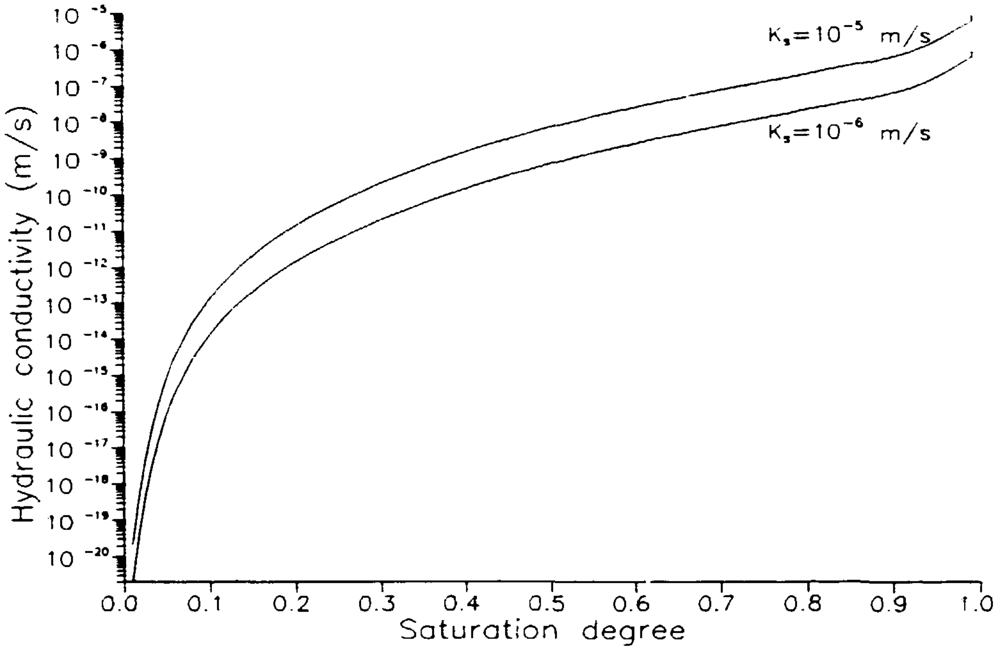

Figure 2.3 *The hydraulic conductivity as a function of saturation degree.*

The two different climatic conditions results in two variations of the top boundary condition, see Figures 2.4 and 2.5. In both cases the inflow through the top surface is taken as the positive net infiltration, i.e. the precipitation minus the evapotranspiration. The net infiltration is positive during the spring and autumn. The negative net infiltration during the summer is modelled as a water removal from the root zone. The curve in Figure 2.4 refers to data from Ställdalen and can be looked upon as normal conditions. The curve in Figure 2.5 refers to data from Malmslätt and represents dry conditions.

The bottom boundary condition were modelled in three different ways, one with a constant level of the groundwater table at a depth of one meter and the other two with a groundwater table varying with the seasons. The variation of the groundwater has been assumed to have the shape of a triangle wave. The depths and the times for the maxima and minima are based on typical variations measured in Sweden, see Figure 2.6. The position of the groundwater level has been fitted to the meteorological data in order to obtain a plausible flow of water in and out of the groundwater column. An alternative approach would have been to make large scale hydrological calculation of the discharge area. However, such a calculation is not trivial to make and would be outside the scope of the present study.

The calculations were made for a two year period after which a semi-stationary condition was reached, i.e. the results at the end of the year were equal to those at the start of the year.

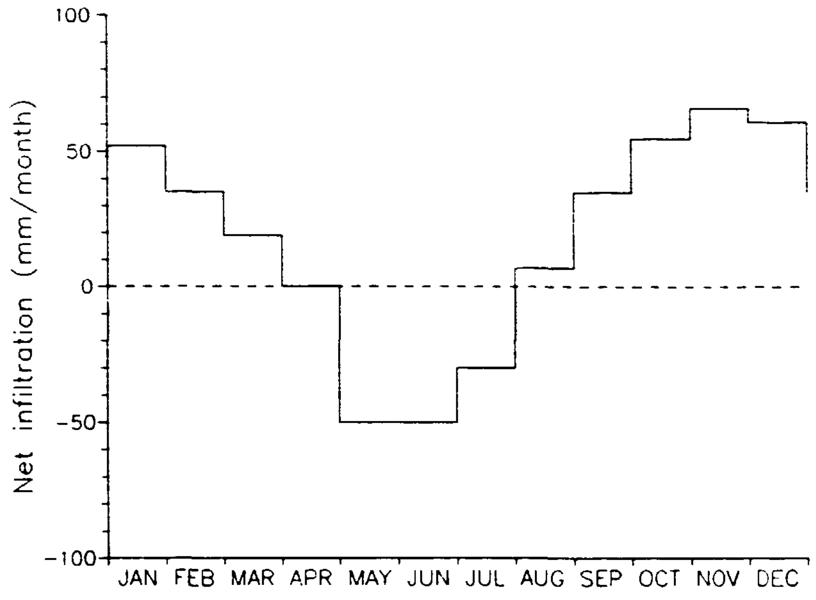

Figure 2.4 ***Variation of top boundary condition in the base case. Taken from net infiltration values from Ställdalen.***

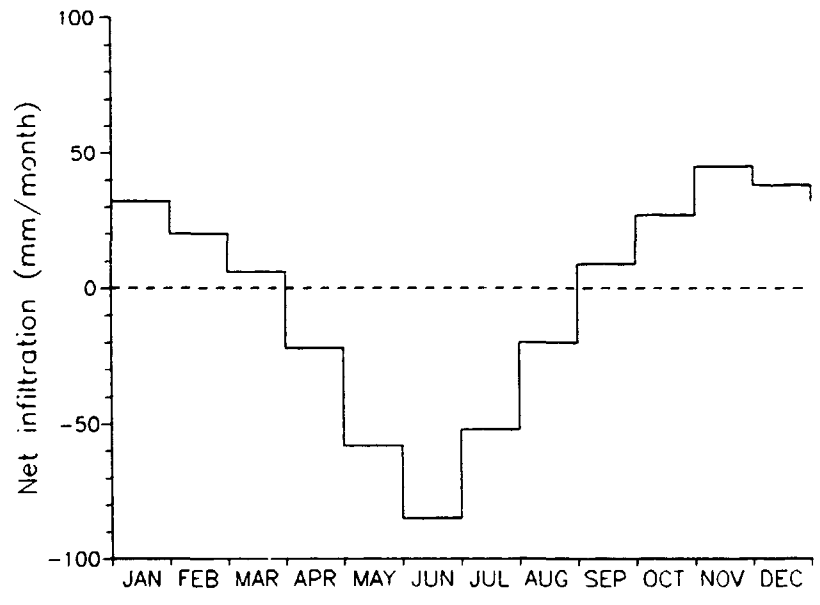

Figure 2.5 ***Variation of top boundary condition for the case with dry conditions, net infiltration taken from Malmslätt.***

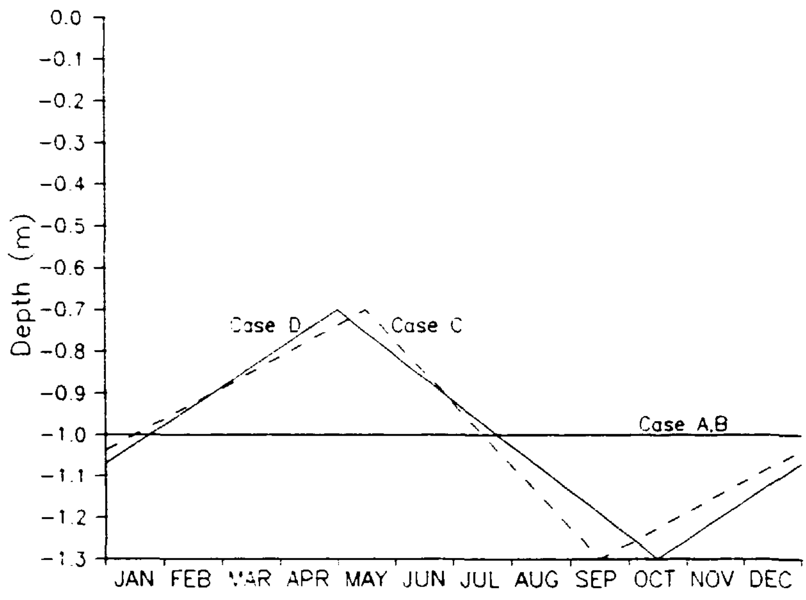

Figure 2.6 *Seasonal variation of groundwater level.*

2.3 Summary of calculation cases

A number of variations concerning the hydrological calculations have been performed where hydraulic conductivity, climate and the groundwater level has been varied, see Table 2.1. The results of the hydrological calculations are presented in Chapter 4.

Table 2.1 *Summary of calculation cases*

Case	Climate	Groundwater level	Material (K_s [m/s])
A6	Normal	Constant	10^{-6}
A5	Normal	Constant	10^{-5}
B6	Dry	Constant	10^{-6}
B5	Dry	Constant	10^{-5}
C6	Normal	Var., max:15/5, min:15/9	10^{-6}
D6	Normal	Var., max:1/5, min:15/10	10^{-6}
D5	Normal	Var., max:1/5, min:15/10	10^{-5}

3 Modelling of radionuclide transport

3.1 Model setup

The radionuclides are assumed to be transported as solutes in the soil water either by advection or by dispersion-diffusion. The transport of the radionuclides is described by the advection-dispersion equation. The computer code TRUMP *[Edwards, 1972]* was used to solve the advection-dispersion equation. The code can take into consideration interaction between solved species and solid surfaces by linear or non-linear sorption. For unsaturated conditions and linear sorption the advection-dispersion equation in one dimension can be written as:

$$(\epsilon S + K_d \rho)\frac{\partial c}{\partial t} = \frac{\partial}{\partial x}\left(D_L(S)\frac{\partial c}{\partial x}\right) - \frac{\partial}{\partial x}(uc) \qquad (3\text{-}1)$$

where c is the solute concentration in the pore water, t is time, x is the distance, ϵ is the porosity, K_d is the sorption coefficient, ρ is the bulk density of the soil, S is the saturation degree, D_L is the dispersion coefficient, and u is the water flow rate. The degree of saturation will affect the capacity term, i.e. the term on the left hand side, and also the dispersion coefficient, D_L.

The water flow rates and the saturation degree from the TRUST calculations are transformed to a form suitable for TRUMP by an automatic procedure *[Collin and Rasmuson, 1990]*. Dispersion can be due to molecular diffusion in the fluid, velocity variations within a flow path or velocity variations between different flow paths. The two last processes are often referred to as hydrodynamic dispersion, which is a complex function of water velocity, saturation and the structure of the material. The dispersion coefficient can be defined as:

$$D_L = \alpha \cdot u + D_0 \cdot S \cdot \epsilon \qquad (3\text{-}2)$$

where:

α is a material specific dispersion length [m]
u is the water flow rate $[m^3\ m^{-2}\ s^{-1}]$
D_0 is the saturated pore diffusivity $[m^2\ s^{-1}]$
S is the saturation degree
ϵ is the porosity.

The first term in the equation describes the hydrodynamic dispersion while the second term describes the molecular diffusion. The automatic procedure has been extended to calculate the dispersivity according to the equation above. By using a constant dispersion length, the dispersion is assumed to increase linearly with the velocity. In the previous study the dispersion was assumed to vary linearly with the degree of saturation, and was described by:

$$D_L = S \cdot D_{LS} \qquad (3\text{-}3)$$

where D_{LS} is the dispersion coefficient for saturated conditions and is derived for an average water flow rate.

3.2 Input data

A collection of experimental evaluations of the dispersion length in unsaturated media has given a range between 1 mm and 0.7 meters *[Gelhar et al. 1985]*. A dispersion length of 0.1 meter was chosen as the base case for this study, with variations within the range 0.05 - 0.4 meters. The saturated pore diffusivity, D_0, has been estimated to $2 \cdot 10^{-10}$ m^2/s. With the water flow rates in the soil obtained in the hydrology calculation it was found that the molecular diffusion will be of minor importance compared to the hydrodynamic dispersion.

Initially the concentration was zero in the whole column. The boundary condition used was a constant concentration of 1 Bq/m^3 in the pore water at the bottom of the modelled soil column, i.e. at 1 meters depth for the cases with constant groundwater level and at 1.3 meters depth for the cases with varying groundwater level. No removal of radionuclides was assumed to occur at the soil surface or due to uptake by plants.

The sorption coefficients, K_d-values, were varied in order to study the influence of different sorption behavior. In the base case a K_d-value of 0 was used to simulate the

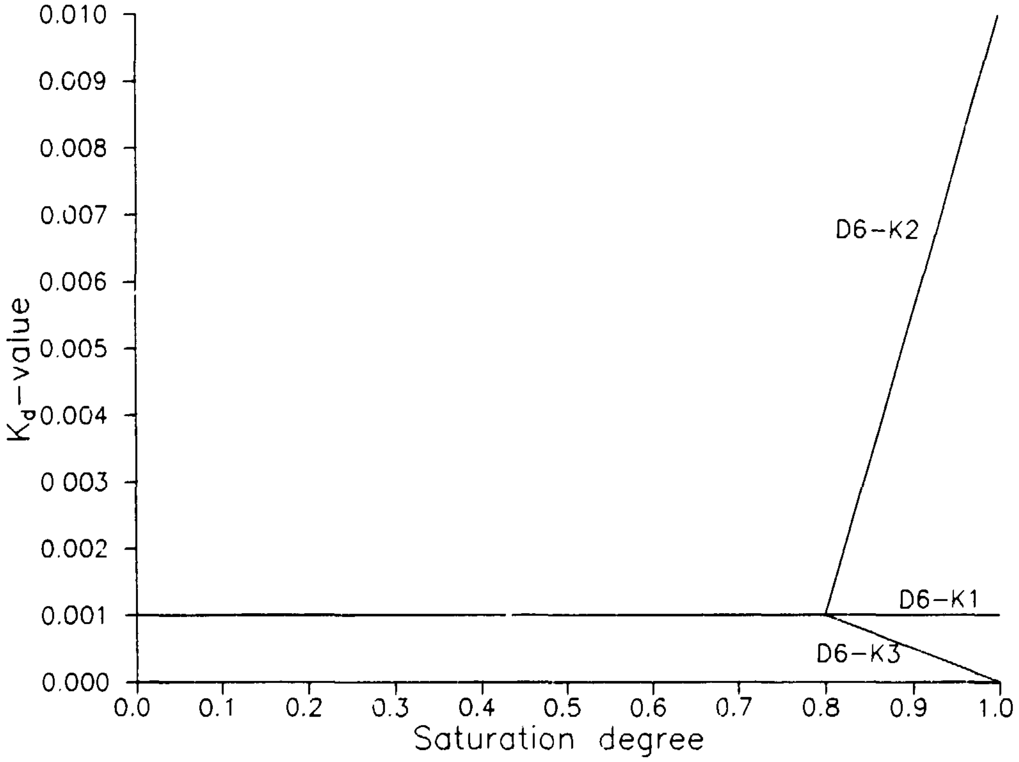

Figure 3.1 *Variation of K_d-value with saturation degree for the three cases used in the calculations.*

transport of a non-sorbing radionuclide. In the variations, a K_d dependent of the redox conditions was simulated by using a K_d dependent of the saturation degree. The redox potential will decrease in a fully saturated soil. A reduction in redox potential leads to an increased sorption for many radionuclides, e.g. neptunium and uranium. However, an increase in redox potential may lead to precipitation of iron, which is an effective sorbent. Thus, an increase in redox potential could potentially result in an increased sorption. Both possibilities were investigated.

Three cases were simulated one with a constant sorption independent of the degree of saturation (K1), a second with a low sorption under unsaturated conditions and a higher sorption under more saturated conditions (K2) and a third case with no sorption under saturated conditions and a slight sorption under unsaturated conditions (K3). The dependencies used are illustrated in Figure 3.1. In the first case, Case D6-K1, a constant K_d of 0.001 m^3/kg was used. In the second case, D6-K2, a K_d of 0.001 m^3/kg was used for saturation degrees below 0.8 and above that a linearly increasing upto 0.01 m^3/kg at full saturation and in the last case, D6-K3, a K_d of 0.001 m^3/kg was used for saturation degrees below 0.8 and above that a linearly decreasing value down to zero at full saturation.

The calculations were continued until a semi-stationary condition was reached in the soil column, i.e. the concentration profile at the end of a year was equal to that at the beginning of the same year. This limited the size of the K_d-values that could be used, due to the long times needed to reach the semi-stationary condition. This takes roughly 5 years assuming no sorption and about 80 years with the K_d-values used in Case D6-K1.

3.3 Summary of calculation cases

A number of transport calculations have been performed varying the dispersion length, the hydrological conditions, the groundwater level and the dispersion in the saturated part of the soil, see Table 3.1. The results of the transport calculations are given in Chapter 4.

Variation of dispersion

For a case with constant saturation and water flow, the two methods described in Section 3.1 for estimation of the dispersion coefficient can be directly compared. However, this cannot be done for a case with varying hydrological conditions, since both water flow and saturation will vary in time and space. Thus each method has been compared with a case with constant hydrological conditions.

Variation of hydrological conditions

The effect on the radionuclide transport of varying the hydrological conditions has been studied by using the water flow rates and saturation data for the corresponding variations performed with the hydrological model. That is variation of the hydraulic conductivity (10^{-6} m/s and 10^{-5} m/s) and variation of climate.

Varying groundwater level

The effect of a time-varying groundwater level on the radionuclide transport using the data from the hydrological cases defined in Chapter 2. In the unsaturated zone the water flow is assumed to be largely vertical and can well be described by the one-dimensional hydrology calculations performed. However, when varying the groundwater level with time, parts of the soil column will be saturated. In this part also a substantial horizontal flow may occur. The magnitude of this flow will depend on the groundwater gradient, which is determined by the local hydrology. This horizontal flow will be of minor importance for the radionuclide transport as long as there are no horizontal concentration gradients. This situation could be expected if the release of radionuclides occurs over a greater area.

In the modelled case, the source of the radionuclides is assumed to be located at a constant depth below the groundwater table. Thus, vertical concentration gradients may occur in the saturated part of the modelled soil column. The horizontal groundwater flow will not only give rise to a hydrodynamic dispersion in the flow direction, but also to a hydrodynamic dispersion perpendicular to the flow direction, so called transversal dispersion. This dispersion will reduce the concentration gradient in the saturated zone. This transversal dispersion may be of importance since it will give a higher concentration at the groundwater level and thus an increased transport of radionuclides to the unsaturated part of the soil.

In order to take this effect into account, the dispersion in the saturated zone has been increased. The additional dispersion is based on a transversal dispersion length of 0.01 meters and a horizontal water flow in the saturated part of $2 \cdot 10^{-7}$ m/s.

Table 3.1 *Summary of transport calculation cases.*

Case	α	Comments
A6-A1	0.05	Smaller dispersion length
A6-R	0.1	Reference case for constant groundwater level
A6-A2	0.2	Two times longer dispersion length
A6-A3	0.4	Four times longer dispersion length
A5-R	0.1	Higher hydraulic conductivity (10^{-5})
B6-R	0.1	Dryer climate
B5-R	0.1	Dryer climate and higher hydraulic conductivity
C6-R	0.1	Varying groundwater level
D6-R	0.1	Reference case for varying groundwater level
D5-R	0.1	Varying groundwater level + higher hydraulic conductivity
D6-M	0.1	Increased disp. in saturated parts
D5-M	0.1	Increased disp. in saturated parts + higher hyd. cond.
D6-K1	0.1	Varying groundwater level, K_d=0.001
D6-K2	0.1	Varying groundwater level, K_d=0.001/0.01
D6-K3	0.1	Varying groundwater level, K_d=0.001/0.0

4 Results

This chapter presents the results of the hydrological and radionuclide transport calculations for the various cases studied. The results of the hydrological calculations will be presented as:

- water saturation profiles in the soil
- water flow rate through the bottom of the soil column

The results of the radionuclide transport calculations will be presented as:

- bulk radionuclide concentration in the root zone (concentration per total volume in the upper 0.3 m of the soil)
- radionuclide concentration in the pore water of the soil

The results of the calculations are presented in graphical form in Figures 4.1 - 4.16. A summary of the average concentration in the pore water and the bulk concentration in the upper 0.3 meters of the soil is presented in Table 4.1.

In the first set of variations, the transport of a non-sorbing radionuclide has been modelled. The reasons for choosing a non-sorbing radionuclide are, firstly, that the effect of changes in the hydrological conditions will become more transparent, and secondly, that the calculation effort will be smaller, since the time needed to reach quasi-stationary conditions is shorter. The results of the previous study [*Elert et al., 1990*] showed that the main difference between a non-sorbing radionuclide and a sorbing radionuclide with constant sorption coefficient was that the bulk concentration was higher for the sorbing radionuclide and that the variation of bulk concentration with time is less prominent. In this study, radionuclide sorption has been included in the last variation where the effect of varying chemical conditions was studied (see Section 4.5).

4.1 Variation of dispersion

The results obtained using a velocity dependent dispersion cannot be directly compared with the results obtained assuming a saturation dependent dispersion, since the two parameters are not readily converted to each other for the case with varying hydrological conditions. However, an estimation of the effect can be obtained by comparing the results of the two cases with the results obtained for a case with a stationary hydrology. That is a stationary downward water flow in the soil, a stationary saturation profile and a constant dispersion coefficient. This stationary case corresponds to Case C of the previous study [*Elert et al., 1990*].

Saturation dependent dispersion

Figure 4.1 shows the bulk concentration in the root zone as a function of the dispersion coefficient for the case with a stationary hydrology and the case with varying hydrology assuming a saturation dependent dispersion. The dispersion coefficient is assumed to be a function of the saturation degree as given in Equation 3-3. For low values of D_L, the case with varying hydrology gives a higher bulk concentration than the case with a stationary hydrology. This is likely due to the dispersive effect obtained by the alternating direction of the water flow in the case of a varying hydrology. This additional dispersion will increase the effective upward transport of radionuclides. For higher values of the dispersion coefficient, the case with the stationary hydrology gives the higher values. An explanation for this may be that in the case of variable hydrology the upward water flow occurs during the summer when the degree of saturation in the soil is lower, thereby giving a lower dispersivity.

Velocity dependent dispersion

In Figure 4.2, the bulk concentration in the root zone is given as a function of the dispersion length for the case with a stationary hydrology and the case with varying hydrology assuming a velocity dependent dispersion. Here the case with a varying hydrology gives a higher bulk concentration in the root zone for all values of the dispersion length. The difference can be attributed to the "dispersive" effect of the alternating direction of the water flow.

The comparisons indicate that the velocity dependent dispersion predicts a more effective upward transport of radionuclides to the root zone. A velocity dependent dispersion is more in accordance with the generally used theories concerning the nature of the dispersion [*Bear, 1972*] and has therefore been assumed for the calculations throughout the rest of the report.

The variation in bulk concentration in the root zone over the year is shown in Figure 4.3 for 4 different values of the dispersion length. It can be seen that the maximum concentration will occur in the late autumn and the lowest values in April. The maximum bulk concentration in the root zone can be about twice the average bulk concentration over the year.

4.2 Variation of hydraulic conductivity

Hydrology

Figure 4.4 shows the water flow rate in and out of the bottom of the soil column for Cases A6 and A5, i.e. with a hydraulic conductivity of 10^{-6} and 10^{-5} m/s, respectively. During the summer and early fall an upward flow from the groundwater to the soil will prevail due to capillary suction from the root zone. During the rest of the year the flow will be directed downward as a result of infiltrating precipitation. The water saturation profile in the soil for the two cases is shown in Figures 4.5a and 4.5b, respectively.

The variation of the hydraulic conductivity will have an important effect on the response time of the water in the soil. For Case A5, with the higher hydraulic conductivity, the response is so fast that the monthly changes in the step function used as boundary condition at the top surface will give an immediate response far down in the soil. This may introduce the need for a more detailed description of the infiltration sequence.

Radionuclide transport

The case with the higher hydraulic conductivity in the soil (Case A5-R) gives a 5 times higher average bulk concentration in the root zone than Case A6-R, 0.020 and 0.004 Bq/m^3, respectively. This is due to the higher upward flow during the summer. Another difference is that the maximum bulk concentration in the root zone occurs earlier in the year, see Figure 4.6.

4.3 Variation of climate

Hydrology

The two different climatic conditions studied gives different hydrological conditions in the soil. In the dryer climate (Cases B5 and B6), the annual evapotranspiration is greater than the annual precipitation, which leads to an average upward flow of water in the soil column. The water flow rate through the bottom of the modelled soil column is shown in Figure 4.7. In the case with the dry climate the inflow will occur during a longer period of time and will in the case with a higher hydraulic conductivity, Case B5, be considerably higher, than what is obtained using the base climate, Case A6.

The water saturation profile will also be affected by the dryer climate, see Figure 4.8. The upper part of the soil profile will become very dry during the summer months. This will have a large impact on the concentration in the pore water.

Radionuclide transport

In Figures 4.9 and 4.10 the concentration in the pore water is shown as a function of depth for the base case climate (Case A6-R) and the case with the dryer climate (Case B6-R), respectively. For the base case climate the concentration in the pore water declines with increasing depth during most months, see Figure 4.9. This is because the annual averaged soil water movement is directed downwards. The upward transport of radionuclides is due to the upward flow during the summer and various dispersion effects. However, the downward transport will be more efficient and thus the concentration in the pore water will be lower in the top part of the soil column. During the summer months there will be large evapotranspiration giving a drying out of the upper soil layer. In the model it is assumed that the removal of radionuclides due to uptake by plants is so small that it can be neglected. Thus, the lowering of the water saturation during the summer will lead to an increase in concentration in the pore water due to evaporation of water.

For the case with a dry climate there will be a net upward transport of water during the year. In average the concentration in the pore water will be higher than assuming the base climate, see Figure 4.10. During the summer the evapotranspiration will lead to a substantial increase in concentration in the pore water. The concentrations may even exceed those in the groundwater. During periods of high infiltration, the concentration in the pore water near the soil surface will decline, due to dilution with rainwater.

For the case with the lower hydraulic conductivity, Case B6-R, the average bulk concentration in the root zone will be roughly 11 times higher for the dryer climate compared to the base climate. If a higher hydraulic conductivity is assumed the dryer climate (Case B5-R) will give roughly 14 times higher bulk radionuclide concentration in the root zone compared to the case with the base climate and the high conductivity, (Case A5-R). The variation of the bulk concentration in the root zone with time is shown in Figure 4.11 (Cases A6-R and B6-R). It can be seen that the variation in climate influences the magnitude of the bulk concentration, but does not significantly change the time for the maximum.

4.4 Time varying groundwater level

The previous cases have been calculated assuming a constant groundwater level 1 meter below the ground surface. However, an alternative model assumption would include also a variation in groundwater level over the year, see Chapter 2.

Hydrology

The following hydrological calculations have been made:

- Case C6 with a maximum groundwater level in the middle of May and a minimum in the middle of September. Hydraulic conductivity 10^{-6} m/s.

- Case D6 with a maximum groundwater level in the beginning of May and a minimum in the middle of October. Hydraulic conductivity 10^{-6} m/s.

- Case D5 as Case D6, but with a hydraulic conductivity of 10^{-5} m/s.

Figure 4.12 shows the calculated flow of water in and out of the bottom of the studied soil column as a function of time for the different cases. A sharp peak can be noticed in the flow for Case C6 at the middle of May and a second peak in the beginning of October. The first peak appears since a net evapotranspiration starts in the beginning of May while the groundwater level is still rising. The inflow is due both to a rising of the groundwater level as a result of water that have infiltrated in other areas further upstream, as well as the onset of a net evapotranspiration. The peak in the autumn is caused by the rise in the groundwater level at the same time as there is a deficit of water in the upper parts of the soil which will give rise to capillary suction. The small adjustment in the times of the maximum and minimum in groundwater level made in Case D6 removes these peaks in the water flow rate, see Figure 4.12.

For the case with the high hydraulic conductivity, Case D5, rapid changes in the water flow rate occurs at the beginning of some months, similar to those obtained in the case with a high hydraulic conductivity and a constant groundwater level. These are also due to the stepwise changes in the boundary condition at the top surface.

The hydrological calculations show that the shape of the curve giving the flow of water from the groundwater to the unsaturated soil column is very sensitive to the combination of infiltration-evapotranspiration data and the groundwater level. The infiltration and evapotranspiration are affected by the conditions at the place where the radionuclide transport is studied, while the groundwater level is affected by the conditions in the whole catchment. We have here chosen to study the radionuclides released to a discharge area located at the bottom of a slope. The groundwater level at the bottom of the slope will be governed by the hydrological conditions over the entire slope. There will be a delay before water infiltrating further up the slope reaches the bottom of the slope and there causes a rise in the groundwater level. This delay will determine the time for maximum and minimum in groundwater level. The extent of this delay is influenced by the size and shape of the slope, hydraulic conductivity of the materials, etc. Thus it is not possible to give a general description of the situation in a given discharge area. The examples presented here can be seen as illustrations of possible effects.

Figure 4.13 gives the water saturation profile at different times for Case D6. The variation in groundwater level can be seen as a larger or smaller part of the soil column having a saturation equal to one. The saturation in the upper part of the soil column is, however, very similar to that obtained in the case with constant groundwater level, compare Figure 4.5a).

Radionuclide transport

The results of the radionuclide transport calculations are presented as bulk concentration in the root zone as a function of time, Figure 4.14. The boundary condition for the radionuclide transport calculations is a constant concentration of 1 Bq/m^3 in the pore water at the bottom of the soil column (depth 1.3 m). This gives a longer transport distance to the root zone of the soil compared to the cases with a constant groundwater level. During the year, the groundwater level varies between 0 and 0.6 meters above the bottom of the soil column. When the groundwater level is high. the radionuclide concentration in the pore water at the groundwater surface may be lower than the 1 Bq/m^3 in the bottom part of the soil column. This contributes to a lower bulk concentration in root zone for the cases assuming a varying groundwater level than in the case with a constant groundwater level. The bulk concentrations obtained using a varying groundwater level are roughly a tenth of the values obtained with a constant level.

The horizontal groundwater flow will give rise to a dispersion perpendicular to the flow direction, transversal dispersion. This dispersion will smear out the concentration profile in the saturated lower part of the soil column and thereby contribute to the upward transport of radionuclides. A schematic description of the dispersion in the saturated zone is given in Figure 4.15. The transversal dispersion is considered in Cases D6-M and D5-M.

The inclusion of the transversal dispersion in the saturated zone has a major effect on the bulk concentration obtained in the root zone. For the cases with the lower hydraulic conductivity, the average bulk concentration in the root zone is increased by almost a factor of 10 when the transversal dispersion is considered, cf. Cases D6-R and D6-M. For Case D5-M (hydraulic conductivity 10^{-5} m/s), the inclusion of transversal dispersion in the saturated zone gives an increase of the bulk concentration in the root zone by almost a factor of 5, compared to the corresponding case without additional dispersion in the saturated zone, Case D5-M.

For the case with varying groundwater level and a lower hydraulic conductivity, the inclusion of transversal dispersion gives an average bulk concentration in the root zone that is similar to that obtained assuming a groundwater level at a constant depth of 1 meter (cf. Cases D6-M and A6-R Figure 4.14). In the case with the higher conductivity the average bulk concentration in the root zone is only half that obtained assuming a constant groundwater level (cf. Cases D5-M and A5-R Figure 4.14). The reason for this is that in the case with a higher conductivity (A5-R) the flow through the bottom of the soil column is to a larger extent determined by the variation of the groundwater surface. Thus, there will be a considerable downward flow of water in the bottom of the soil column during the early autumn, while there is an upward flow during the same period for the case with a lower hydraulic conductivity (A6-R).

4.5 Variation of sorption

The effect of redox dependent sorption has been studied for a case with varying hydrology and a varying groundwater level. A low sorbing radionuclide was chosen in order to get reasonable computation times with the model. The distribution coefficient (Kd-value) for the radionuclide is assumed to be a function of the redox potential, which in turn is dependent on the level of water saturation. Thus, the Kd-value is given as a function of the saturation, see Section 3.2.

For case D6-K1 (constant K_d=0.001 m^3/kg), the average bulk concentration in the root zone is roughly three times higher than for the corresponding case with no sorption (Case D6-R). This difference is due to the larger storage capacity in the root zone for sorbing radionuclides and does not indicate an increased transport of radionuclides to the root zone. In the case with a higher sorption in the saturated part (Case D6-K2), the average bulk concentration in the root zone is roughly four times higher than in the case with no sorption and 20% higher than in the case with constant sorption. Also in this case is the difference mainly due to the larger storage capacity for radionuclides in the root zone. The very small increase compared to the case with constant sorption is probably due to the somewhat higher sorption during the months when the water saturation in the root zone is slightly above 0.8, see Figure 4.13. The variation of the bulk concentration in the root zone over the year is small, see Figure 4.16. This is due to the large storage capacity for radionuclides in the root zone. Also the concentration in the pore water of the root zone is nearly constant, the major variation will occur in the part of the soil column where the groundwater level fluctuates.

The highest average bulk concentration in the root zone is obtained for the case with a K_d of 0.001 up to a saturation degree of 0.8, Case D6-K3. The value is roughly 5.5 times higher than for the case with no sorption at all.

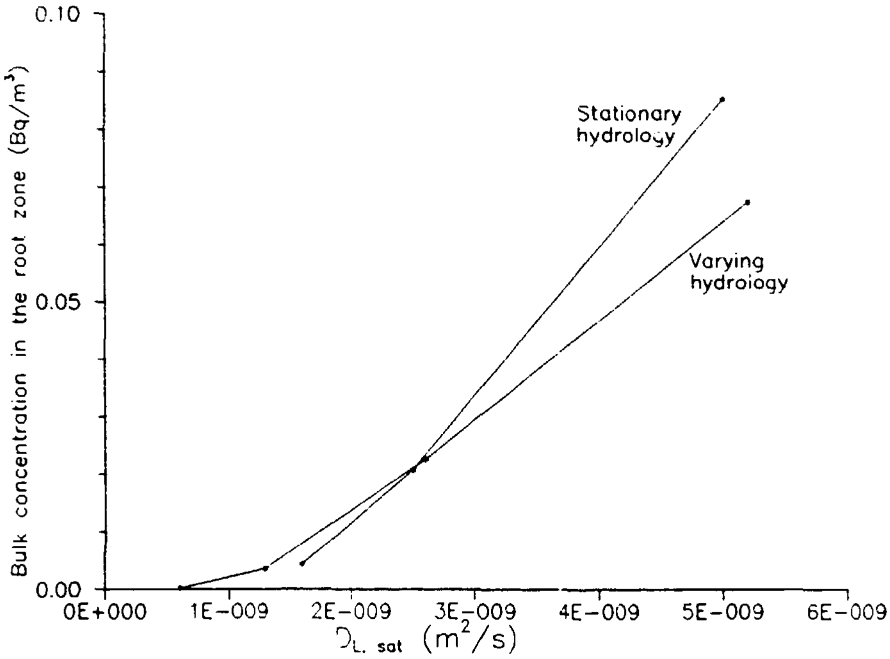

Figure 4.1 ***Bulk concentration in root zone as a function of the dispersion coefficient for the case with stationary hydrology and the case with varying hydrology.***

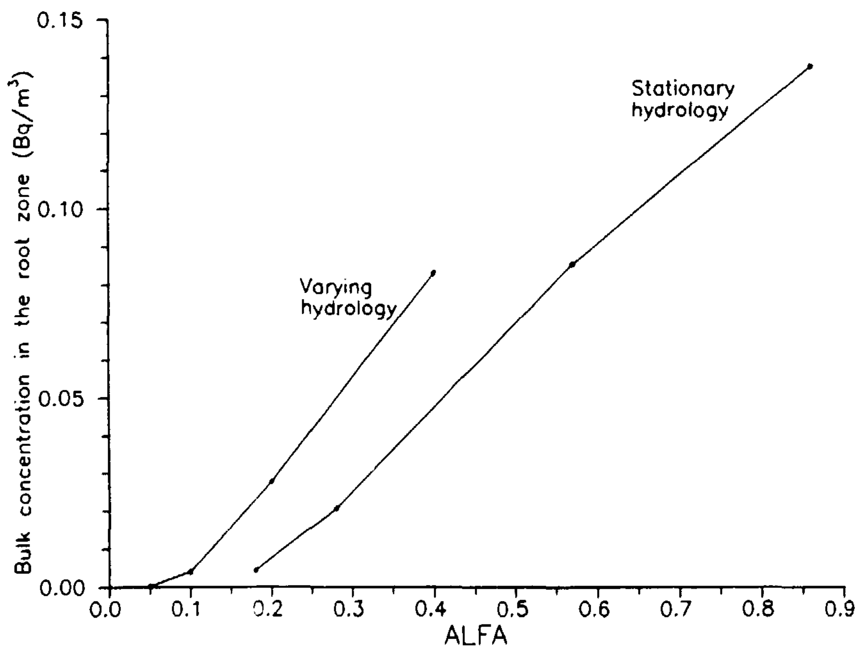

Figure 4.2 ***Bulk concentration in the root zone as a function of the dispersion length, α, for the case with stationary hydrology and the case with varying hydrology.***

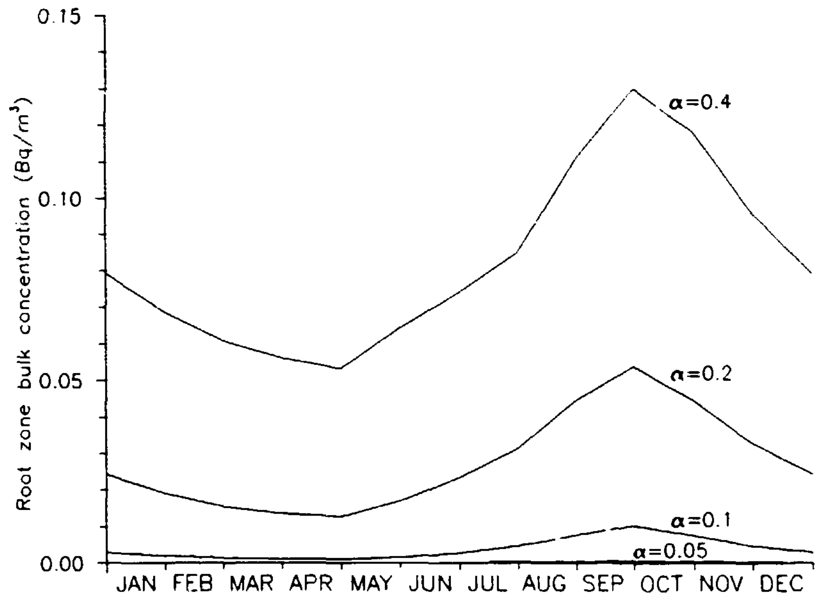

Figure 4.3 **Variation in bulk concentration in the root zone over the year for 4 different values of the dispersion length, α.**

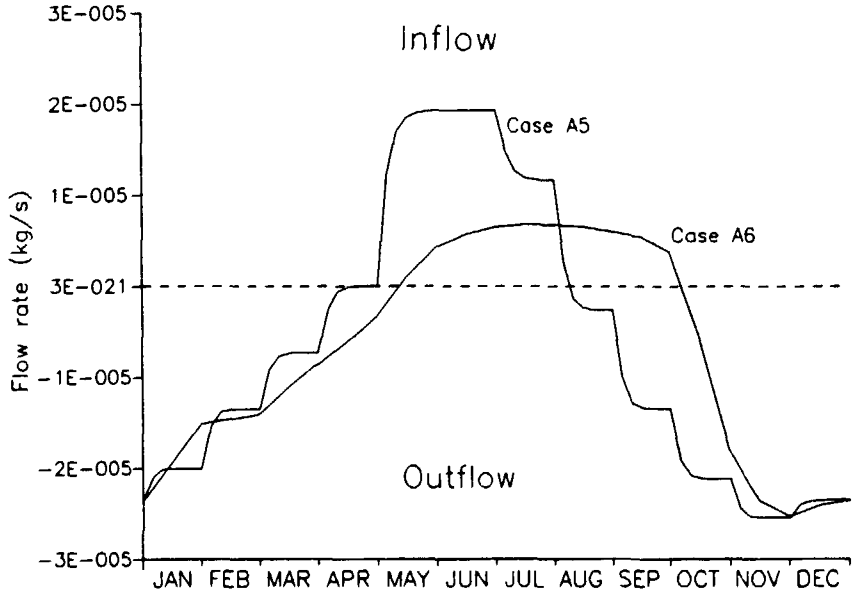

Figure 4.4 **Water flow rate through the bottom of the soil column for Case A6 and Case A5.**

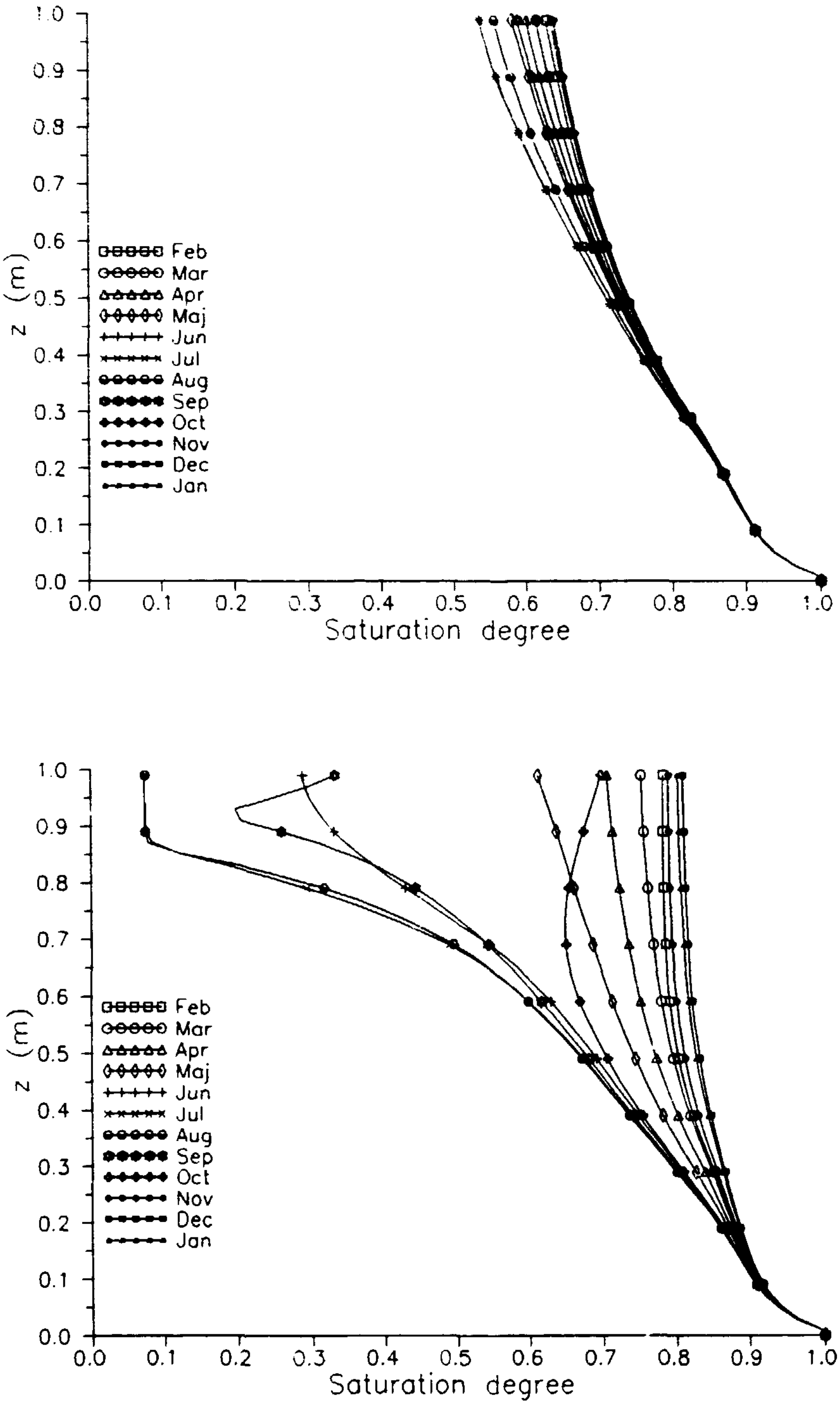

Figure 4.5 *Water saturation as a function of depth for Case A6 (upper) and A5 (lower).*

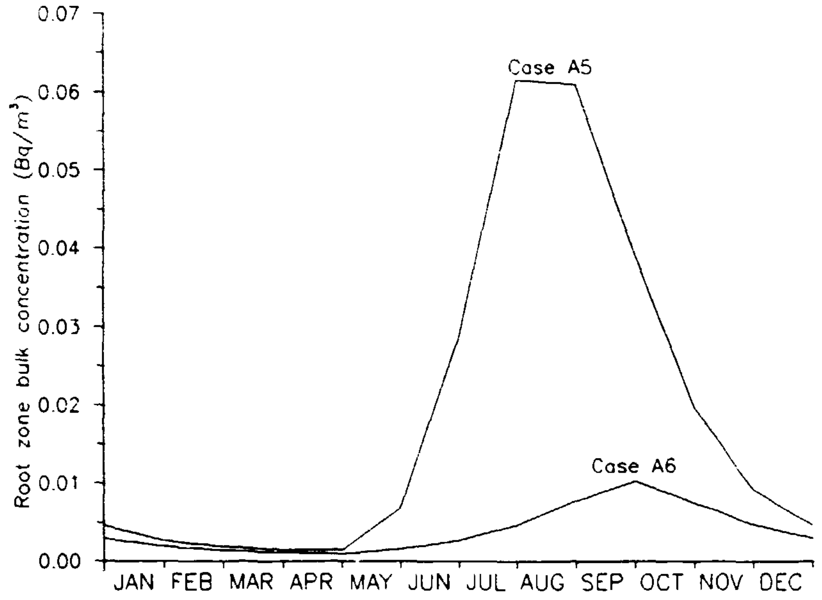

Figure 4.6 ***Variation in bulk concentration in the root zone over the year for Case A6 and Case A5.***

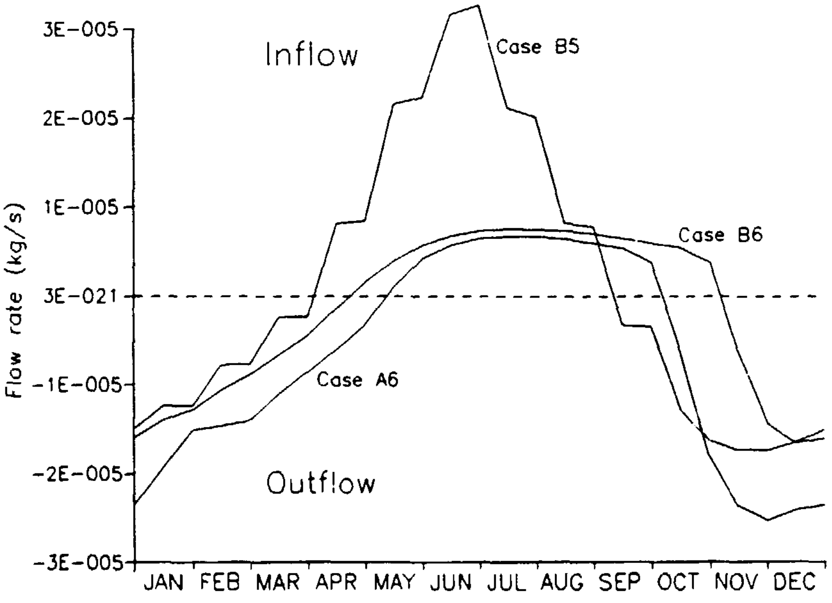

Figure 4.7 ***Water flow rate through the bottom of the soil column for Case A6 (wet climate $K=10^{-6}$ m/s), Case B6 (dry climate $K=10^{-6}$ m/s), and Case B5 (dry climate $K=10^{-5}$ m/s).***

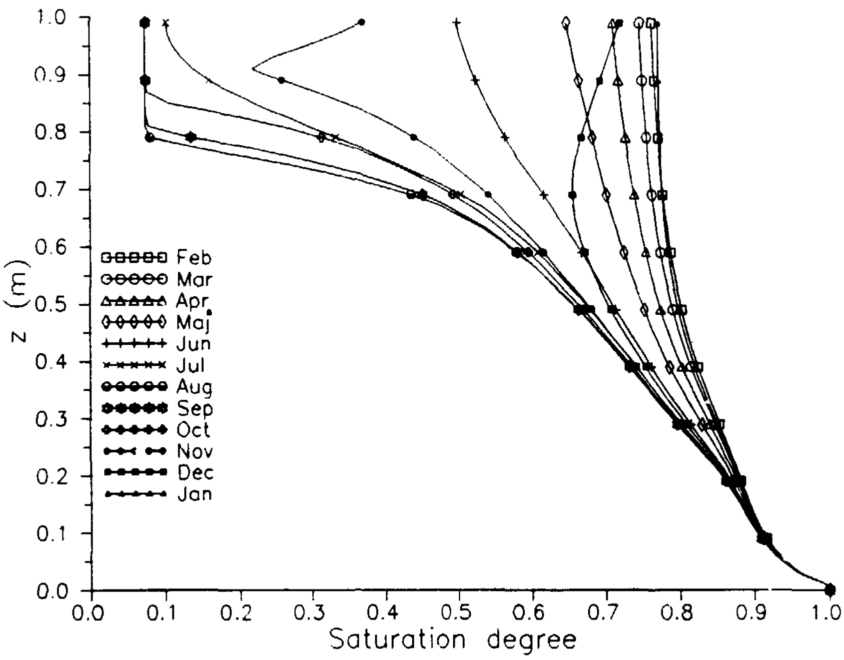

Figure 4.8 ***Water saturation as a function of depth for case with dry climate (B6).***

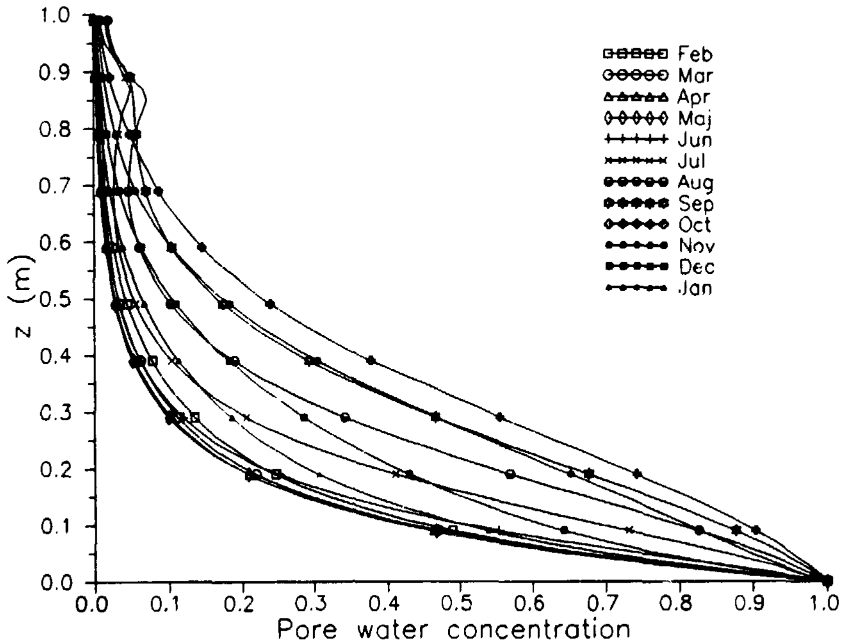

Figure 4.9 ***Concentration in pore water as a function of depth for Case A6-R.***

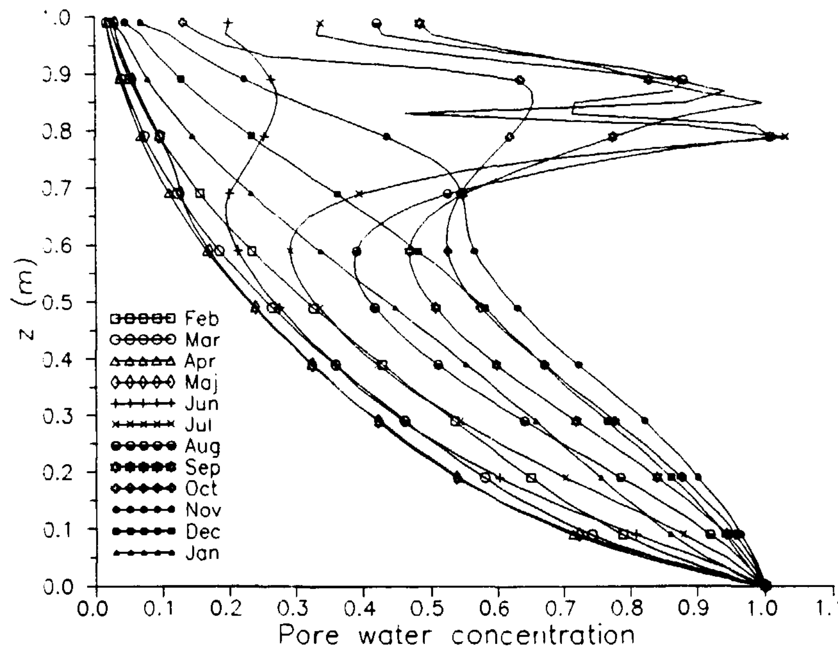

Figure 4.10 *Concentration in pore water as a function of depth for Case B6-R.*

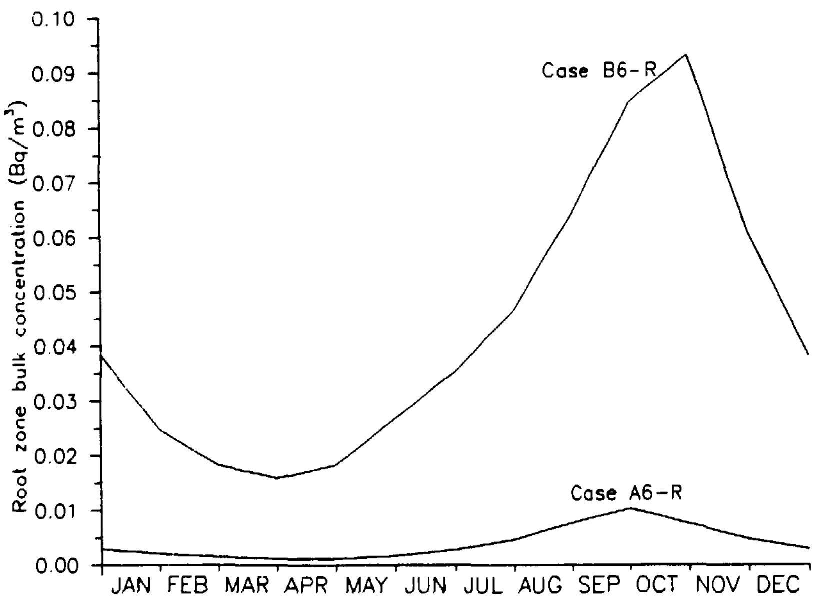

Figure 4.11 *Variation in bulk concentration in the root zone over the year for cases with different climate (Case A6-R and Case B6-R).*

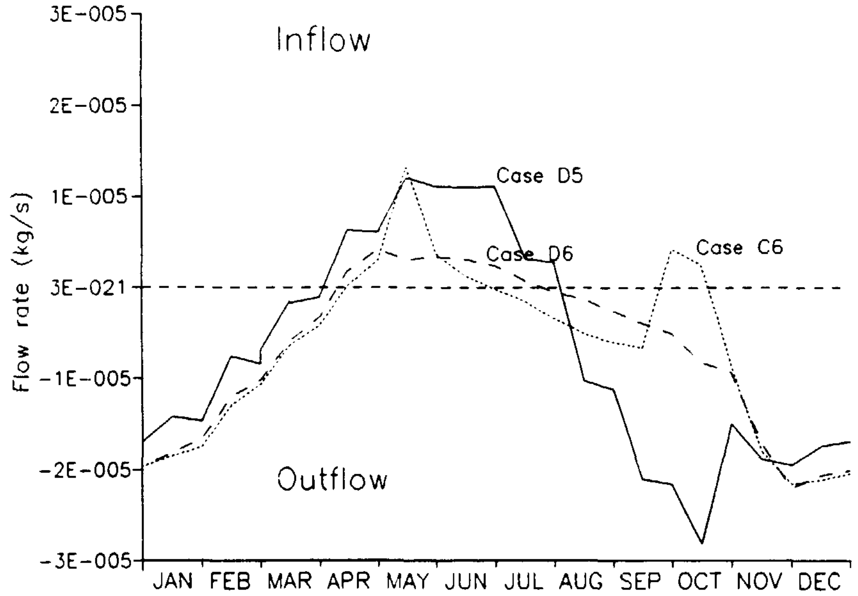

Figure 4.12 ***Water flow rate through the bottom of the soil column for cases with a varying groundwater level (Case C6, Case D6 and Case D5).***

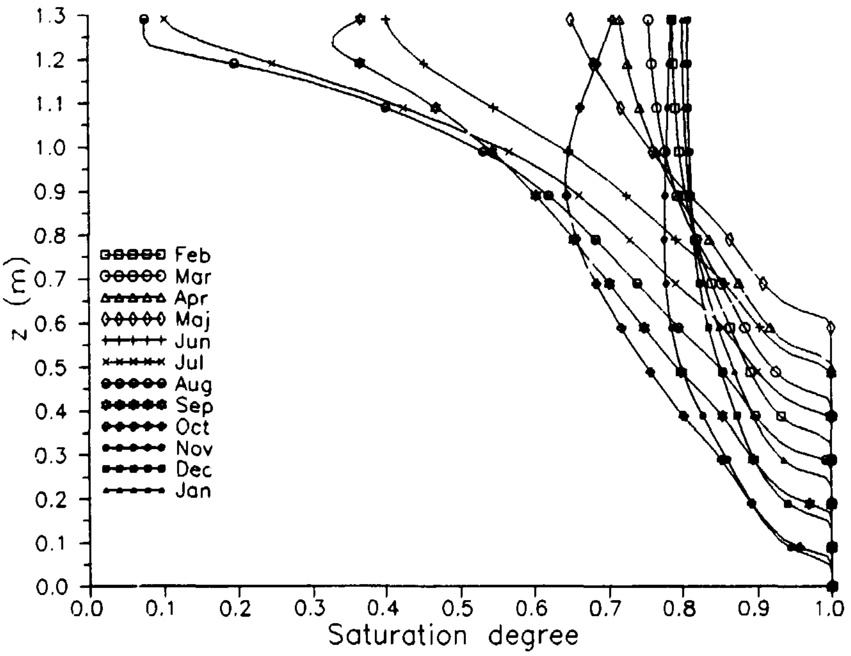

Figure 4.13 ***Water saturation as a funciion of depth for Case D6.***

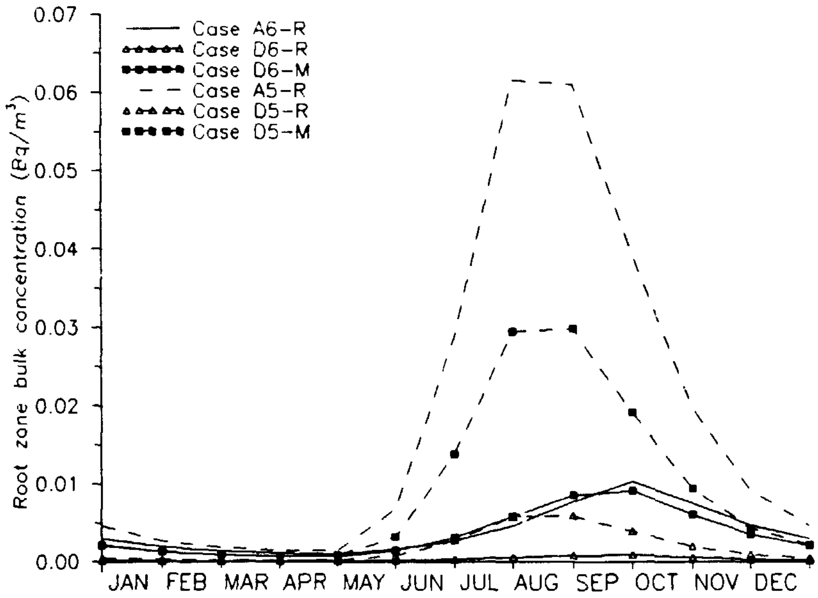

Figure 4.14 *Variation in bulk concentration in the root zone over the year for cases with varying groundwater level, Cases D6-R and D5-R without transversal dispersion, Cases D6-M and D5-R with transversal dispersion. Cases A6-R and A5-R with constant groundwater level added as a comparison.*

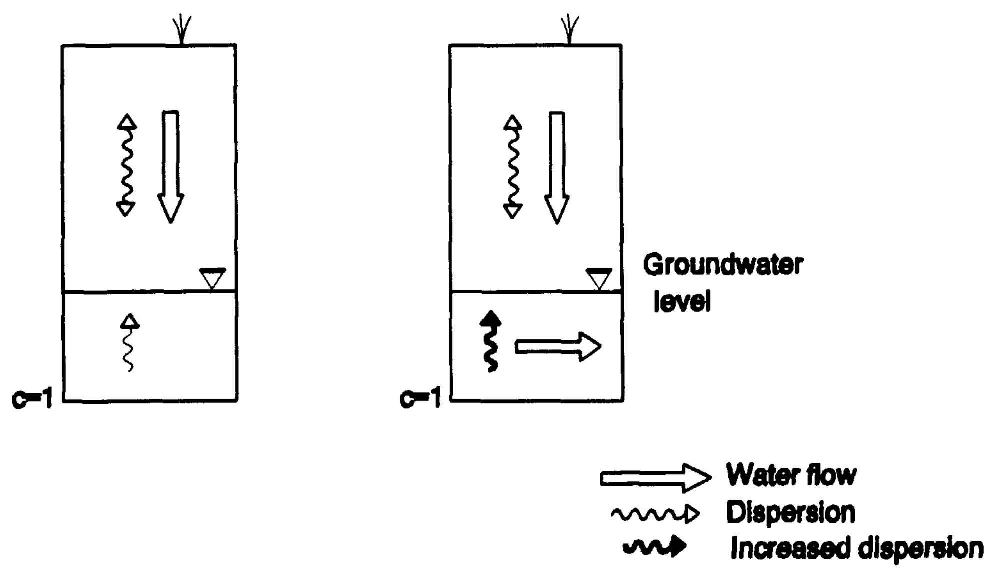

Figure 4.15 *Schematic description of the effect of the dispersion in the saturated part of the soil column.*

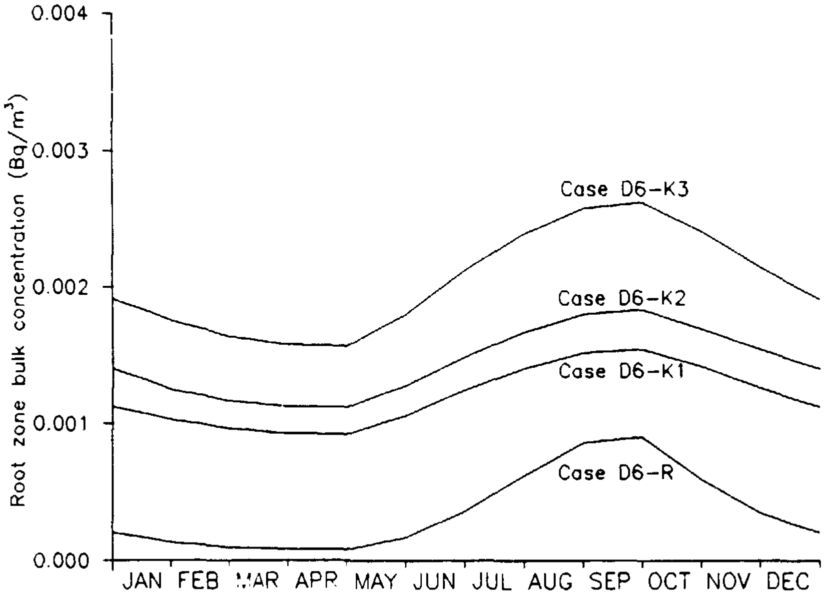

Figure 4.16 **Variation in bulk concentration in the root zone over the year for cases with varying groundwater level and K_d**

Table 4.1 **Summary of the calculated annual average radionuclide concentration in the pore water and the bulk soil of the root zone (upper 0.3 meters).**

Case	Pore water Bq/m^3	Bulk conc. Bq/m^3
A5-R	0.072	0.0200
A6-R	0.019	0.0040
B5-R	1.105	0.2900
B6-R	0.309	0.0440
D5-R	0.007	0.0020
D6-R	0.002	0.0004
D5-M	0.035	0.0096
D6-M	0.016	0.0036

5 Estimation of effective parameters

5.1 Comparison with box model calculations

Biosphere transport modelling is often made with compartment or box models. The different components of the biosphere are represented as a series of compartments where the rate of radionuclide transport between the compartments is the product of a transfer factor and the radionuclide amount in the "donor" compartment. Inside each compartment an instantaneous and complete mixing is assumed. The transfer factors may be empirically derived or estimated based on simple submodels. The transport of radionuclides in soils is usually modelled with two or three compartments representing different layers of the soil, e.g. subsoil and top soil and the transfer factors are estimated by simple submodels. The main transport process is usually advection with the soil water, combined with linear sorption. Other processes such as erosion, bioturbation and diffusion may also be included in the submodels. The transfer factor due to advection can be written as:

$$k_{12} = u_{12}/M_1 \qquad (5\text{-}1)$$

where:

k_{12} is the transfer factor between compartments 1 and 2
u_{12} is the water flow rate between compartments 1 and 2
M_1 is the amount of water in compartment 1

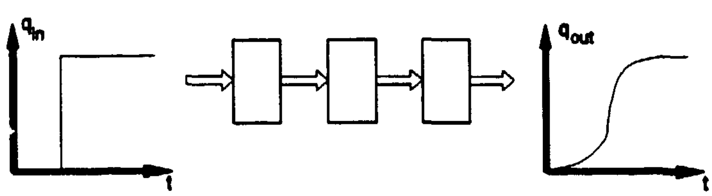

Figure 5.1 Response of a sharp pulse passing a series of compartments.

The assumption of a complete mixing within the compartments will give rise to dispersion of the radionuclide transported through the series of compartments. A sharp pulse is entered at one end will give a spread out release in the other end, see Figure 5.1. The degree of dispersion will depend on the dimension of the compartments, a few large compartments will give rise to a large dispersion while a large number of small compartments will give rise to a small dispersion. In order to compare the behaviour of the advection-dispersion model and the compartment model a method to equate the description of the dispersion must be derived.

5.1.1 Derivation of dispersion values

The transport through a soil column can be characterized by the residence time distribution (RTD) for the solutes in the column, see Figure 5.2. A residence time distribution may be derived from experiments or estimated from simulations with mathematical models.

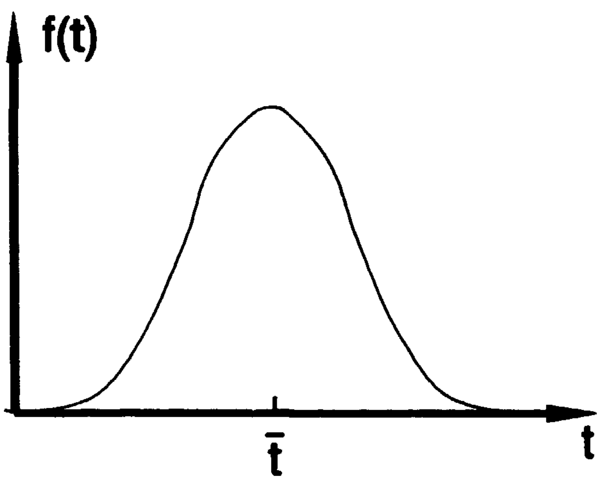

Figure 5.2 Typical residence time distribution (RTD).

The residence time distribution can be characterized by its mean, $\bar{t}$, and variance σ^2, where the variance is a measure of the dispersion in the column. For the advection-dispersion model the variance at the outlet may be written as a function of the dispersion coefficient and the water flow rate if the variance of the tracer pulse at the inlet is known. The difference in variance is then given as [*Levenspiel, 1972*]:

$$\frac{\Delta\sigma^2}{\bar{t}^2} = \frac{\sigma^2_{out} - \sigma^2_{in}}{\bar{t}^2} = 2\frac{D_L}{UX} \tag{5-2}$$

where:

$\bar{t}$ is the mean of the residence time distribution
σ^2 is the variance of the residence time distribution
X is the length of the column in the flow direction
U is the water flow rate
D_L is the dispersion coefficient

If the input variance is zero as with a pulse or step input, the expression can be simplified as:

$$\frac{\sigma^2}{\bar{t}^2} = 2\frac{D_L}{UX} \tag{5-3}$$

For a series of N equal-size compartments the mean residence time for the series is the sum of the mean residence time of the individual compartments:

$$\bar{t} = N\,\bar{t}_i \tag{5-4}$$

and the variance is the sum of the square of the mean residence time for the compartments:

$$\sigma^2 = N\,\bar{t}_i^2 = \frac{\bar{t}^2}{N} \tag{5-5}$$

Using Equation 5-4 and 5-5 an expression for the "inherent" dispersion coefficient as a function of the number of compartments or the length of the compartments can be derived.

$$D_L = \frac{1}{2}\frac{UX}{N} = \frac{1}{2}U\Delta x \tag{5-6}$$

U in Equation 5-6 corresponds to the total flow from a compartment. In the presently studied case, there will be an upward water flow in the soil due to capillary rise during periods with excess evapotranspiration. Additionally, there will be a downward water flow in the soil due to infiltrating precipitation. Thus in our case, U in Equation 5-6 is the sum of the downward flow and the upward water flow, U_{tot}.

The "inherent" dispersion coefficient will be proportional to the size of the compartments or inversely proportional to the number of compartments. The "inherent" dispersion will also increase with the water flow within the soil. This is of importance since the inherent dispersion will have a strong influence on the concentration of radionuclides in the root zone.

In the steady-state situation, the upward transport, by advection and by dispersion, will balance the downward transport, and a constant radionuclide concentration profile will be obtained in the soil. The concentration profile in the soil at this stationary state can easily be evaluated by solving the advection-dispersion equation. This gives:

$$c(x) = c_0 e^{-x\frac{U_{diff}}{D_L}} \tag{5-7}$$

where:

c_0 is the concentration in the groundwater at the bottom of the soil column
U_{diff} is the net downward water flow in the soil, $u_{down} - u_{up}$.
x is the distance from the groundwater surface

5.1.2 Comparison between compartment model and advection-dispersion model

The results obtained with the steady-state advection-dispersion model has been compared with results obtained with a simple steady-state compartment model using annual averages of water flow.

In the simple compartment model, the soil has been divided into a number of equal-size compartments. The upward transfer factor was determined from the annual net evapotranspiration (88 mm/a) and the downward transfer factor was determined from the annual net infiltration (298 mm/a). Thus, there is "simultaneously" a flow directed downward and upward. In the first calculations only the "inherent" dispersion obtained by the assumption of mixed compartments was included. A schematic description of the set of compartments is given in Figure 5.3. Figure 5.4 shows the concentration in the pore water as a function of height above the groundwater level obtained with different number of compartments. It is clear that the number of compartments has a large influence on the radionuclide concentration in the upper part of the soil column. A large number of compartments will give a small dispersion and a low concentration in the upper parts of the soil. Thus, the modelling approach chosen will have an important effect on the results obtained.

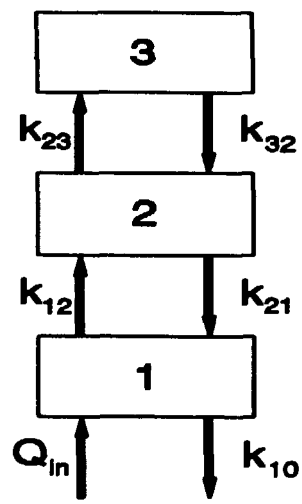

Figure 5.3 Schematic picture of a set of compartments.

In order to test the derivation of dispersion values from Section 5.1.1, a comparison was made with a simple analytical solution of the advection-dispersion equation assuming a downward flow of radionuclides and an upward dispersive transport. The dispersion coefficient used in the analytical model was chosen to give matching values of the radionuclide concentration in the root zone for the model using 20 compartments ($D_L = 0.01$ m^2/a). The dispersion coefficient was then increased by a factors of 2, 4, and 10, in order to correspond with the "inherent" dispersion obtained using 10, 5, and 2 compartments, respectively. Figure 5.5 shows that there is a good correspondence between the results of the compartment model and the advection-dispersion model for all discretizations. The deviation in the results from the two models is largest in the part of the soil immediately above the groundwater surface. This is due to a slight inconsistency between the boundary conditions for the two models.

The variation of the "inherent" dispersivity with rate of water flow within the soil has also been studied. A comparison was made between the "fitted" dispersivity coefficients giving equivalent results with the advection-dispersion model and the compartment model, and the dispersion coefficient derived using Equation 5-6. 20 boxes were used in the compartment model. Both the flow U_{tot}, given as the sum of the upward and downward flow and the difference between the downward flow and the upward flow, U_{diff} were varied. Figure 5.6 shows that the "fitted" dispersivity coefficients are slightly lower than those derived by the equation. The deviation from the theoretical value is largest when the difference between the upward and downward flow is large.

5.1.3 Derivation of effective dispersion parameters

In the previous section it is shown that the number of compartment has a strong influence on the degree of dispersion obtained in the system. A biosphere modeller using a compartment model describing an advective-dispersive processes should ideally adapt the number of compartment to the amount of dispersion that can be expected to occur in the system. However, for various reasons this may not be practically achieved, e.g. the number of compartments required may be excessively large or may not conform with other requirements the modeller may have. It would thus be beneficial if the modeller could adapt the transfer coefficients to obtain the desired degree of dispersion with an arbitrary division of the system into compartments. Below follows an outline of for such a procedure when modelling the upward transport in soils.

The compartment model of a soil system consists of boxes with bi-directional flow. In a wet environment the annual downward water flow will be larger that the annual upward water flow. For such a case the ratio of the amount of radionuclides in two connected compartments will equal the ratio between the upward and downward flow between the two compartments i and j:

$$\frac{Q_i}{Q_j} = \frac{k_{ji}}{k_{ij}} \tag{5-8}$$

where:

Q_i is the total activity in compartment i

k_{ij} is the transfer factor between compartments i and j

If compartment j is placed above compartment i and the annual downward and upward is constant throughout the soil column (i.e. $k_{ij} = k_{jk}$) the total activity in the n:th compartment will be:

$$Q_n = \left(\frac{k_{ij}}{k_{ji}}\right)^n \cdot Q_1 \tag{5-9}$$

When a large number of compartments is used to describe the soil transport and the resulting dispersion is lower than the desired, an additional dispersion term may be added to the transfer factor as given in Equation 5-1. The dispersive term may be defined as:

$$k_{ij}^{D} = \frac{2D^{*} \cdot A_{ij}}{M_i(\Delta x_i + \Delta x_j)} \tag{5-10}$$

k_{ij}^{D} is the dispersive transfer factor between compartments i and j
D^{*} is an effective dispersivity coefficient taking considering the dispersion not taken into account by the "inherent" dispersion
A_{ij} is the connection area between compartments i and j
M_i is the amount of water in compartment i
Δx_i is the width of compartment i

For equal size compartments $k_{ij}^{D} = k_{ji}^{D}$. By adding a term to the transfer factors the ratio k_{ij}/k_{ji} will be reduced. The result will be an increased activity content in the upper part of the soil column at stationary conditions. The main problem with this approach is to find generally applicable methods to derive D^{*}. The value of D^{*} should be such that the residence time distributions of the compartment model should correspond to the expected one in the modelled system. For the case many compartments are used this could be done by matching the variance of the RTD. One method to do this would be to estimate the "inherent" dispersion coefficient using Equation 5-6 and then add the amount needed to reach the desired dispersion coefficient.

In principle the same methodology should be applicable also for a case where only a limited number of compartments are used to model the soil column and the "inherent" dispersion obtained is higher than the desired dispersion. In this case the transfer factors should be reduced by a given amount. However, the correction procedures described above will have some limitations. For examples, there may be difficulties in having a reasonable correction if only a limited number of compartments are used (two or three), since the residence time distribution of the compartment model will become skewed if the number of compartments is small. Thus, there may be large differences in the RTD although the variances are matched.

5.2 Comparison with the cases evaluated with TRUST-TRUMP

A simple compartment model has been applied to the cases evaluated with TRUST-TRUMP. The annual downward and upward water flow from the hydrological calculations were used to derive transfer factors in a simple 2-box compartment model. The steady-state concentrations in the root zone were evaluated and the results were compared with the results obtained in the numerical calculations. In Table 5.1 the results obtained with the different methods are compiled. For the cases with a constant groundwater level (Cases A5-R, A6-R, B5-R and B6-R), the compartment model gives a 1.5 - 3 times higher bulk concentration in the root zone than the annual average obtained with TRUST-TRUMP. In the case with a varying groundwater level the compartment model gives a more than ten times higher bulk concentration than TRUST-TRUMP for the Cases D5-R and D6-R. This may be due to the fact that the TRUST-TRUMP values are low due to the problems with the boundary conditions, see Section 4.4. The difference between the two models is smaller for the cases where the transverse dispersion has been added (Cases D5-M and D6-M).

The comparison between the two models shows that the compartment model in all cases predicts a higher radionuclide concentration in the root zone than the complex advection-dispersion model. However, the magnitude of the difference in the predictions is small compared to other error sources. It should be noted that these predictions have been made only for a very limited set of soil conditions.

Table 5.1 *Comparison between radionuclide concentrations in the root zone obtained with TRUMP/TRUST and a simple compartment model.*

Case	TRUST/TRUMP Cpore Bq/m^3	TRUST/TRUMP Cbulk Bq/m^3	Box model Cpore Bq/m^3	Box model Cbulk Bq/m^3	Cbox/Ctrump -
A5-R	0.072	0.0200	0.1420	0.0395	1.97
A6-R	0.019	0.0040	0.0541	0.0114	2.85
B5-R	1.105	0.2900	1.6119	0.4230	1.46
B6-R	0.309	0.0440	0.4167	0.0593	1.35
D5-R	0.007	0.0020	0.0929	0.0258	12.9
D6-R	0.002	0.0004	0.0182	0.0040	10.7
D5-M	0.035	0.0096	0.0929	0.0255	2.65
D6-M	0.016	0.0036	0.0182	0.0041	1.14

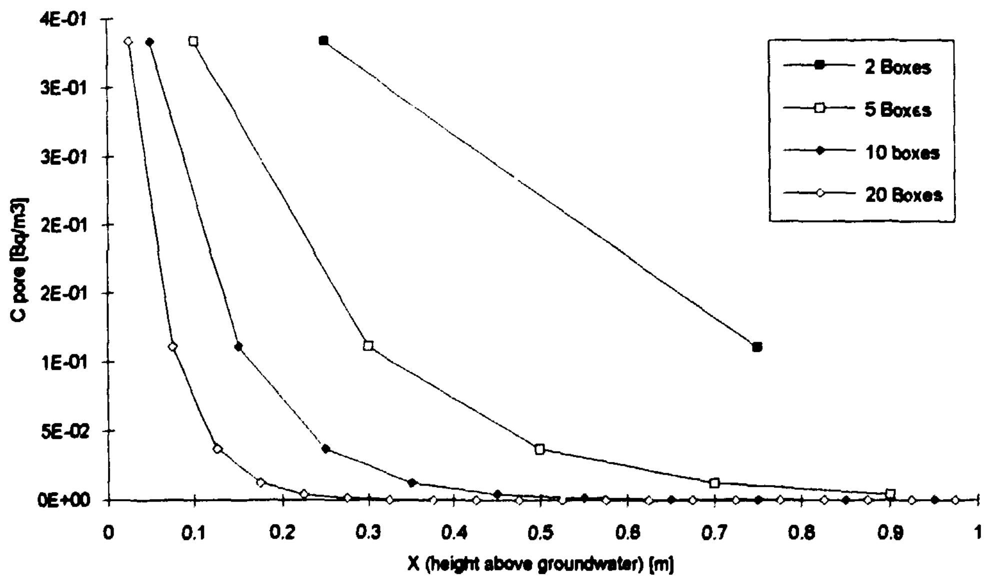

Figure 5.4 Concentration in the pore water as a function of the height above the groundwater table. Calculated with a box model with 2, 5, 10 and 20 boxes.

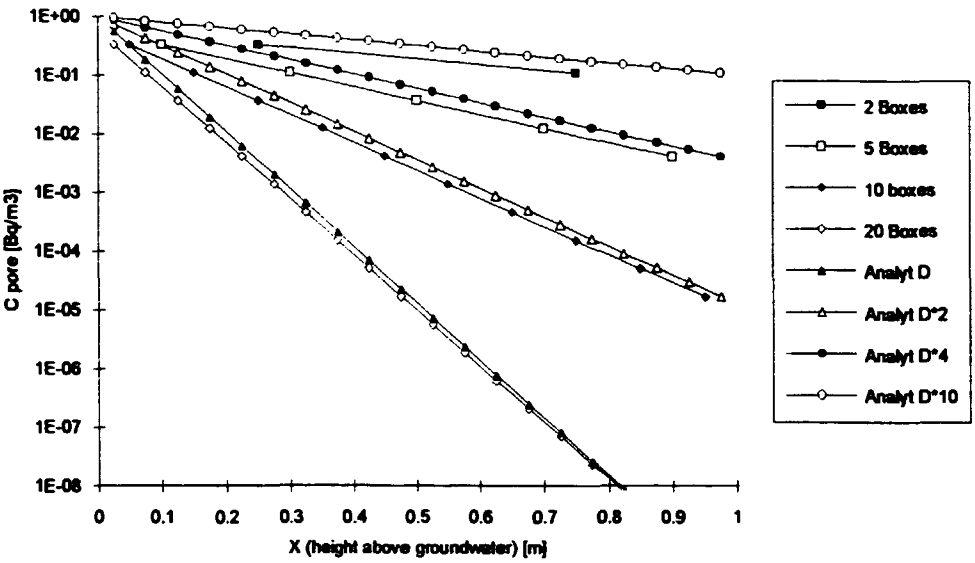

Figure 5.5 Concentration profile in pore water calculated with compartment model and advection-dispersion model with equivalent dispersivity.

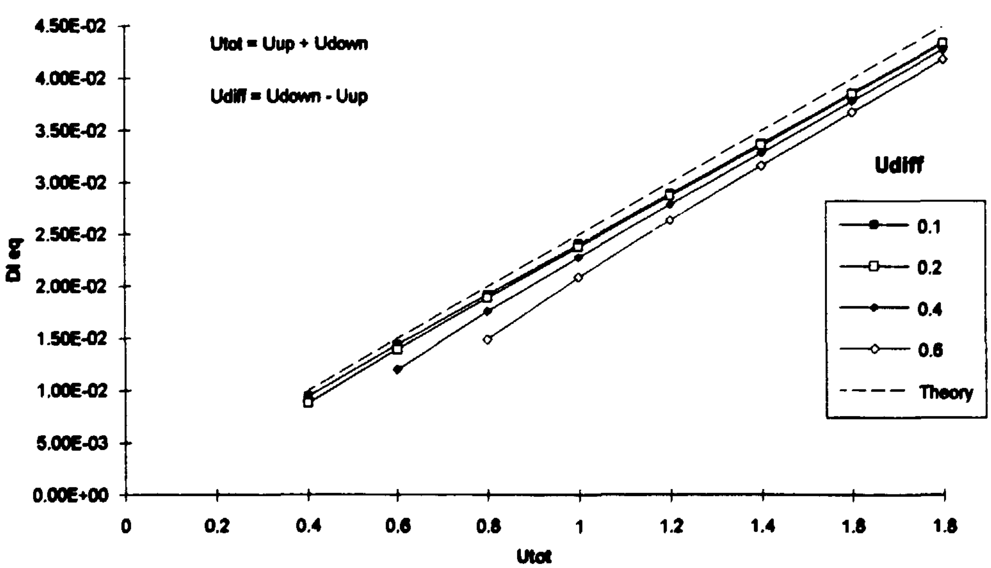

Figure 5.6 Dispersivity coefficients giving equivalent results with advection dispersion model and compartment model.

6 Discussion and conclusions

Processes for radionuclide transport in soils

This study has focused on the transport of dissolved radionuclides in the soil water by advection and dispersion. However, also other processes such as soil erosion, bioturbation and transport with colloids may be of importance.

The soil water movement is largely determined by the climatic conditions. Two main cases of climatic conditions can be identified. The first case corresponds to a wet climate case with an excess in precipitation as compared to evapotranspiration, giving annual net downward flow of soil water. The second case corresponds to a dry climate with an excess of evapotranspiration as compared to precipitation, with a net upward flow of soil water as a result.

For the wet climate case the main process for transporting radionuclides from the groundwater level to the upper parts of the soil is the upward transport that will prevail over parts of the year. Additionally, radionuclides can be transported by molecular diffusion. In the modelling it may not be practical to consider very short term fluctuations in precipitation and evapotranspiration, instead the water movement is averaged over days, weeks, months or even annually. Thus, the transport of radionuclides caused by short term fluctuations must be considered in a different way. In this study we have chosen to consider the transport due to short term fluctuations as a dispersive process. Dispersion is a composite process that is caused by large and small scale variations in the water flow velocity. These velocity variations may in turn be caused by heterogeneities in the soil. For climates with large surplus precipitation, the upward transport will not be very efficient. Thus, the radionuclide concentration in pore water in the root zone will be considerably lower than in the groundwater.

For the dry climate there will be a net upward flow of soil water. This leads to an efficient upward transport of radionuclides in the soil. Since water evaporates in the upper parts of the soil column there may be a further increase in the radionuclide concentration in the pore water. In the case of a large evapotranspiration, the concentration in the pore water of the root zone may even exceed the concentration in the groundwater. Dispersion and diffusion may then give rise to a downward transport of radionuclides, since dispersion and diffusion will give a radionuclide transport in opposite direction of the concentration gradient.

This study has mainly been concerned with the wet climate conditions, since these are most frequent in Sweden. A variation of the climate conditions has been performed, see below.

Modelling of dispersion

The dispersion parameter is important when modelling the upward radionuclide transport in a wet climate. The method to include dispersion in the model depends on the type of model used, as well as on the scale and the type of soil system modelled.

Furthermore, the dependencies of the dispersion parameter on saturation and water velocity in an unsaturated system can be defined in several ways. In this study two alternative methodologies were used: one with dispersion mainly dependent on saturation and one with dispersion mainly dependent on water velocity. The results of the calculations show that the velocity dependent dispersion leads to a more effective upward transport of radionuclides to the root zone. The velocity dependent dispersion was used for the rest of the calculations since it is in accordance with the generally used theories concerning the nature of dispersion.

Variations of hydraulic conductivity

Variations of the hydraulic conductivity of the soil showed that an increase in soil conductivity leads to an increase in the radionuclide concentration in the root zone. The main reason is that the varying boundary condition at the surface (i.e. the effect of precipitation and evapotranspiration) will have an effect on the soil water movement deeper in the soil column if the hydraulic conductivity is higher. Thus the exchange of water between the soil and the groundwater will increase giving a higher inflow of radionuclides during periods of large evapotranspiration. However, the results of the hydrological calculations with the higher hydraulic conductivity ($K = 10^{-5}$ m/s) suggest that an averaging of precipitation and evapotranspiration over monthly periods is too long. A shorter integration interval would increase the exchange of water between the soil and the groundwater, and may thus predict a larger upward radionuclide transport.

Variation of climatic conditions

The climatic conditions (i.e. the precipitation and evapotranspiration) will have a large influence on the rate of upward transport of radionuclides as described above. In the present study only two climatic conditions have been studied a "wet" and a "dry" climate. The annual average root zone bulk concentration was more than 10 times higher with the dry climate than with the wet climate. The two climates studied were of the same principle type, with excess precipitation during spring and autumn, and with a dry summer period. The difference between the two climate types was mainly the amount of precipitation and evapotranspiration. Other climate types of interest may be where part of the winter precipitation comes as snow on a frozen soil. The soil water movement will then be restricted during the winter and a large part of the precipitation may be removed as surface run-off during the snow-melt. Consequently the downward soil water movement will be reduced, which in turn can lead to increased radionuclide concentrations in the top soil.

Variations of groundwater level

Calculations have also been performed where a seasonal variation in the groundwater level has been simulated. In a real situation there will be a connection between the precipitation and evapotranspiration and the groundwater movement. However, in order to simulate this, a much larger area needs to be considered. Thus, the movement of the groundwater level was given as an external condition, but it was adapted to fit the meteorological data used.

The first results of the calculations with a varying groundwater level gave a radionuclide concentration in the root zone one tenth of that obtained in the calculations performed with a stationary groundwater level. There may be several reasons for this difference. For the case with a varying groundwater level, the distance between the groundwater and the root zone is greater during the summer when there is an upward soil water flow. This decreases the exchange of water between the soil and the groundwater and increases the length of the transport path for the radionuclides. Secondly, the boundary condition at the bottom is set as a constant concentration in the pore water at the bottom of the soil column. Thus, the concentration in the pore water at the groundwater level may be lower than for the case with a constant groundwater level.

The boundary condition used in the case with a varying groundwater level may be more realistic than the boundary condition applied for the case with a constant groundwater level. For radionuclides coming from an underground repository the source term will not be a constant concentration in the pore water at the groundwater level, but rather a more or less constant rate of inflow of contaminated deep groundwater. Thus can a decrease in concentration at the upper surface of the groundwater be expected during periods with large infiltration and rising groundwater levels because of the higher degree of dilution. More complex calculations in two dimensions could consider also such effects, but would unnecessarily complicate the description of the flow in the unsaturated part of the soil. In this part of the soil the transport can be assumed to be mainly vertical. Thus, we choose to use a simplified description of the flow in the saturated part of the soil and only consider the vertical transport.

The horizontal component of the groundwater flow will give rise to a dispersion perpendicular to the flow direction. This dispersion would give a less sharp concentration profile in the saturated part of the soil column and thereby contribute to the upward transport of radionuclides. The results of the calculations considering this transversal dispersion gave a concentration of radionuclides in the root zone of the same order as those obtained in the calculations with a constant groundwater level

Variation of sorption

The main effect of a varying groundwater level was expected to occur when combined with a variation of sorption. Sorption of many radionuclides is redox sensitive and the redox potential in the soil will be affected by the degree of water saturation. Thus, the degree of sorption can be expected to be affected by the degree of water saturation. This can increase the upward transport of radionuclides if the periods with an upward flow occur when the radionuclide sorption is low and the periods with a downward flow occur when the sorption is high. If the situation is the opposite, a decrease of the upward radionuclide flow may occur. Calculations were performed with a radionuclide sorption given as a function of saturatior These functions were not based on realistic data, since the purpose was only to detect the possible effect of the coupling between saturation and sorption.

The performed calculations did not result in any significant change in the bulk radionuclide concentration in the root zone due to the variation in sorption. As a maximum a 60% higher bulk concentration was obtained compared to the case with a constant sorption. The reason may be that the expected effect does not occur. However, additional explanations may also be possible. For example, the distribution coefficients (Kd-values) were restricted to low values (< 0.01 m^3/kg) in order to keep the computation times reasonable. This may make it difficult to detect any effects of the variation in sorption. Secondly, the functions for how the sorption varies with saturation may have been chosen in such a way that the effect of saturation variations not was apparent.

Further parameters that may be important

There are additional parameters of potential importance apart from those that have been analyzed in this study. One such parameter is the depth of the groundwater table. An increased depth will decrease the exchange of water between the soil and the groundwater as well as increase the migration distance for the radionuclides. Both changes will decrease the radionuclide concentration in the root zone.

Comparison with compartment model

The annual average concentrations in the root zone obtained with the numerical calculations were compared with those obtained with a simple steady-state compartment model. The transfer factors in the compartment model were derived from the results of the hydrological calculations. The comparison shows that the compartment model in all cases gives a higher prediction of the annual average radionuclide concentration in the root zone than the numerical model. The magnitude of the difference varies between a factor of 2 and 10. However, the numerical calculations with a varying hydrology results in large variations in the concentration over the year. During the summer the concentration in the root zone may be several times higher than the annual average. This may be important for plant uptake, since this increased concentration coincides with the plant growing season.

The calculations made with the simple compartment model also show that these types of models have an inherent dispersion that depends on the size of the compartments. This inherent dispersion must be considered when assigning suitable parameters for compartment models. This inherent dispersion may lead to high estimates of the concentration in the root zone if long soil columns are modelled with only a few compartments. On the other hand a large number of compartments may lead to very low estimates of the root zone concentration. Various methods to compensate for this inherent dispersion have been investigated, but additional work remains before any generally applicable method could be derived.

Concluding remarks

As a conclusion it can be stated that the variations in soil hydrology will be of importance for the upward transport of radionuclides. The various cases studied show that the climatic conditions and the hydraulic conductivity of the soil is of importance. In this study, as well as in most other studies, the soil water movement is treated separately from the large scale hydrology of the discharge area. Since in reality they are connected systems, this leads to problems when defining suitable boundary conditions for the radionuclide input. This problem was especially apparent when studying the effect of varying the groundwater level.

The complex models needed to consider a varying soil hydrology are often impractical to use in safety assessments. Instead more simple compartment models are usually preferred. However, the comparisons made between the two types of models show that the set up of the compartment models may have a significant impact on the results obtained. Thus the division into compartments and the choice of input parameters must be made with care. Further comparisons between simple and more complex models may provide valuable insights in how to best perform averaging of time or spatially varying parameters and how to derive effective parameters for use in assessment models.

Notation

A_{ij} connection area between compartments *i* and *j* (m^2)

c solute concentration (Bq/m^3)

c_0 concentration in the groundwater (Bq/m^3)

D_0 saturated pore diffusivity (m^2/s)

D_L dispersion coefficient (m^2/s)

D_{LS} dispersion coefficient for saturated conditions (m^2/s)

D^* effective dispersivity coefficient taking considering the dispersion not taken into account by the "inherent" dispersion (m^2/s)

K_d sorption coefficient (m^3/kg)

K_s saturated hydraulic conductivity (m/s)

k_{ij} transfer factor between compartments *i* and *j* (s^{-1})

k_{ij}^D dispersive transfer factor between compartments *i* and *j* (s^{-1})

M_i amount of water in compartment *i* (m^3)

N number of compartments

Q_i total activity in compartment *i* (Bq)

S saturation degree

t time (yr), (s)

$\bar{t}$ mean of the residence time distribution (s)

u, U water flow rate ($m^3\ m^{-2}\ s^{-1}$)

U_{diff} net downward flow in soil ($m^3\ m^{-2}\ s^{-1}$)

U_{tot} sum of upward and downward flow in soil ($m^3\ m^{-2}\ s^{-1}$)

u_{ij} water flow rate between compartments *i* and *j* (m^3/s)

x distance (m)

X length of the column in the flow direction (m)

Greek letters

α dispersion length (m)

Δx_i width of compartment *i*

ϵ porosity (m^3/m^3)

ρ density (kg/m^3)

σ^2 variance of the residence time distribution

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