



# TECHNICAL REPORT 89-20

## BIOSPHERE MODELLING FOR A DEEP RADIOACTIVE WASTE REPOSITORY: TREATMENT OF THE GROUNDWATER-SOIL PATHWAY

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NOVEMBER 1990

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## **Preface**

In the framework of its Waste Management Programm the Paul Scherrer Institute is performing work to increase the understanding of radionuclide transport in the biosphere. These investigations are performed in close cooperation with, and with the financial support of NAGRA. The present report is issued simultaneously as a PSI report and a NAGRA NTB.

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## Abstract

The effect of radionuclide transfer from near-surface groundwater to the rooting zone soil, via a deep soil layer, is modelled in this report. The possible extent of upward solute movement is evaluated for a region in northern Switzerland.

The concentrations of  $^{237}\text{Np}$  and  $^{129}\text{I}$  in the deep and top soil, and hence growing crops, are evaluated assuming a constant unit activity concentration in the groundwater. A number of parameter variations are considered, namely variable soil sorption coefficients, reduced infiltration of rain water and decreased groundwater flow. A release to an alternative smaller recipient region in northern Switzerland is also evaluated. For the parameter ranges considered uncertainty in the solid-liquid distribution coefficient has the largest effect on overall uncertainty.

These calculations have been presented within the international Biosphere Model Validation Study (BIOMOVS). A description of the test scenario, and the model calculations submitted, have been included in this report for completeness.

To place the groundwater-soil-crop-man pathway in context, its contribution to the total dose to man is evaluated for the  $^{237}\text{Np}$  -  $^{233}\text{U}$  -  $^{229}\text{Th}$  decay chain. The results obtained using the two-layer soil model, described in this report, are compared with the single-layer soil model used during Project Gewähr 1985. The more realistic two-layer soil model indicated an increase in importance of the drinking water pathway. It should be noted, however, that not all the critical pathways have been treated in this study with the same degree of conservatism.

## Résumé

Les effets du transport de radionucléides provenant d'eaux souterraines peu profondes jusqu'à la surface sont modélisés dans ce rapport. Dans ce modèle le sol a été représenté par deux couches, l'une au dessus de la nappe d'eaux (1m), l'autre comprenant la zone des racines (0.25 m). L'ampleur du mouvement ascendant des solutions est évaluée pour une région du nord de la Suisse.

Les concentrations de  $^{237}\text{Np}$  et  $^{129}\text{I}$  dans le sol en profondeur et en surface, et donc dans les cultures, sont calculées pour un cas de référence. Un certain nombre de variations paramétrique est considéré, telle que la réduction de l'infiltration, la diminution du débit des eaux souterraines, la variation de la sorption (retardation) ainsi que la réduction de la zone de collection d'eau dans une plus petite région dans le nord de la Suisse. Parmi les paramètres considérés, c'est la répartition entre les phases solides et liquides dans le sol qui conduit à la plus grande incertitude globale.

Les résultats des calculs du cas de référence et des incertitudes qui lui sont associées, sont également présentés pour le scénario de contrôle B6 (transport d'eaux souterraines contaminées vers le sol) de l'étude internationale de "Biosphere Model Validation Study" (BIOMOVS).

La dose reçue par l'homme par le cheminement eaux souterraines-sol-culture-hommes est évaluée pour la chaîne  $^{237}\text{Np}$  -  $^{233}\text{U}$  -  $^{229}\text{Th}$  en utilisant le terme source du Project Gewähr 1985. Ces résultats sont comparés avec les doses précédemment calculées. Le modèle plus réaliste (sol à deux couches), met en évidence l'importance prédominante de la voie "eau potable". Toutefois, pas tous les cheminements critiques n'ont pas été traités dans cette étude au même degré de conservativité.

## Zusammenfassung

In diesem Bericht werden die Auswirkungen des Radionuklidtransports aus oberflächennahem Grundwasser durch eine untere Bodenschicht bis in den Bereich der Wurzelzone modelliert. Es wird die Bedeutung der aufwärts gerichteten Bewegung gelöster Stoffe für eine Region der Nordschweiz untersucht.

Die Konzentrationen von  $^{237}\text{Np}$  und  $^{129}\text{I}$  in der unteren und oberen Bodenschicht (und damit in den Kulturpflanzen) werden unter der Annahme einer konstanten Einheitsaktivität im Grundwasser für einen Basisfall berechnet. Es werden einige Parameter-Variationen durchgeführt nämlich: reduzierte Infiltration, geringerer Grundwasserfluss und Retardierung (Sorptions), sowie ein alternatives, kleineres Einzugsgebiet in der Nordschweiz. Von den untersuchten Parametern führt die Unsicherheit der Verteilungskoeffizienten zwischen fester und flüssiger Phase im Boden zu den grössten Gesamtunsicherheiten.

Vorgestellt werden auch die Resultate der Basisfall-Rechnungen und die zugehörigen Unsicherheiten für das Test-Szenario B6 (Transport von kontaminiertem Grundwasser in den Boden) der internationalen "Biosphere Model Validation Study" (BIOMOVS).

Der Beitrag des Pfades Grundwasser-Boden-Kulturpflanze-Mensch zur Gesamtdosis wird für die Nuklid-Kette  $^{237}\text{Np}$  -  $^{233}\text{U}$  -  $^{229}\text{Th}$  berechnet, unter Verwendung des Quellterms aus dem Projekt-Gewähr 1985. Das realistischere Modell eines Zweischicht-Bodens zeigte eine zunehmende Bedeutung des Trinkwasserpfades, wobei jedoch in dieser Studie nicht alle kritischen Transportwege mit dem gleichen Grad an Konservativität behandelt wurden.

# 1 Introduction

In the safety analysis of nuclear waste disposal in deep underground geological formations in Switzerland, the most probable pathway identified for radionuclides released from such repositories to reach the biosphere is via groundwater. Consequently, groundwater represents a potentially important exposure pathway for radiation doses to man. In Project Gewähr (NAGRA, 1985), this pathway comprised the base case and direct use of the water for drinking has shown to contribute significantly to doses for most of the nuclides considered if it is assumed to become contaminated.

In the biosphere modelling previously carried out (Bundi, 1984; Grogan, 1985; NAGRA, 1985) it was assumed that the solutes in the groundwater passed directly through the top soil layer. In general, groundwater does not flow directly and entirely into the surface soil, however, this was conservatively assumed for the model calculations in order to assess the radiological consequences. In this report a more realistic situation is evaluated in which only a fraction of the solutes in groundwater pass up to the soil and the remaining part flow through the deep soil into a river. For this reason the soil is divided into a deep soil compartment and a top soil compartment.

In chapter 2 of this report, the conceptual model is described and the water balance in the rooting zone soil evaluated. Although the net transport is downwards in Switzerland (total annual precipitation is greater than the annual evapotranspiration), upward transport during shorter periods of time might result in the upward movement of solutes. The possible extent of upward transport and the evaluation of the waterflows between the different physical compartments is evaluated for the Laufenburg region in northern Switzerland. This region was selected as the base case scenario in the feasibility studies and the safety assessment for the Swiss waste disposal programme (NAGRA, 1985).

Two important parameters for modelling the accumulation of radionuclides in the soil and the subsequent transfer through the foodchain to man are the solid-liquid distribution coefficient ( $K_d$ ) and the soil-to-plant transfer factors. These parameter values are evaluated for two radionuclides,  $^{237}\text{Np}$  and  $^{129}\text{I}$ , and are used in the calculations. Food-chain products are attached to the different physical compartments and their radionuclide concentrations calculated using simple steady-state transfer factors. For the foodcrops attached to the top soil compartment (i.e. pasture, cereals, leaf vegetable and root vegetables), an inverse relationship between the transfer factor and  $K_d$  parameters is proposed.

The hydrological data and parameter values defined in the chapters 2 and 3 are used for the calculations of the test scenario B6 (transport of contaminated groundwater to soil – BIOMOVs, 1987) of the international model intercomparison “BIOMOVs” using the computer code BIOPATH. Parameter variations with respect to reduced infiltration, decreased groundwater flow, compartment sizes and waterfluxes between the compartments and sorption onto both deep and top soil have been carried out. From these results an evaluation is made of the potential extent of contamination of rooting zone soil, root crops growing on this soil and the contamination of the air (as inhaled), due to the presence of contaminated groundwater flow in the deep soil. These results have been presented in the BIOMOVs study for the B6 groundwater scenario.

In chapter 5, the radiation doses to man for the neptunium decay chain are calculated analogous to the way they are presented in Project Gewähr (NAGRA, 1985). By using the same source term, a direct comparison is made between the results and the differences arising from the application of a more realistic near-surface groundwater model are discussed.

## 2 Groundwater model description

### 2.1 The conceptual model

A fundamental difference with the previous models (NAGRA, 1985; Grogan, 1985) is the division of the soil compartment into two layers: a one meter thick deep soil layer and a 0.25 m top soil layer, which is assumed to represent the plant rooting zone. Solutes transported with groundwater (originating, for example, from a waste repository) are assumed to flow through the deep soil compartment. In the region modelled here, the groundwater flows out into a large river. It is assumed that a certain amount of the groundwater can pass up into the upper soil layer resulting in the accumulation of solutes in the rooting zone and hence the growing crops. Such a situation can occur if the capillary fringe intersects the surface so that evapotranspiration (i.e. the combination of evaporation of water from the soil surface and transpiration by the crops) results in an upward movement of solutes present in the groundwater. The upward movement of solutes could also be brought about by changes in the height of the groundwater table (which are seasonal), but this situation is not modelled here. The precipitation falling onto the soil partly infiltrates the deep soil and partly goes as run-off directly to the river.

The system of compartments used to model this situation is presented in Figure 1. This linear system is described by coupled first order differential equations that are solved numerically using the biosphere computer code BIOPATH (Røjder et al. 1988). The radionuclide concentration in each of the compartments is calculated as a function of time. Between the two soil compartments, the contamination flow upwards and the diluting flow downwards are assumed to occur simultaneously, averaged over yearly timesteps. However, in reality, both flows might be temporally separated.

It is clear that this model is only applicable to soils that have a permanent groundwater table that is situated not too far from the growing crops (i.e. at depth of 1 to 2 m) so that as a result of capillary rise the top of the capillary fringe is at the ground surface. The extent of capillary rise is almost entirely determined by the physical properties of the soil such as particle and pore size distributions. For regions where the groundwater table lies at greater depths (more than 3 m) capillary rise is unlikely to influence the upward movement of solutes in water. Soils which are characterized by a very high groundwater table (hydromorphic soils) are very sparse in Switzerland (Müller, 1989)

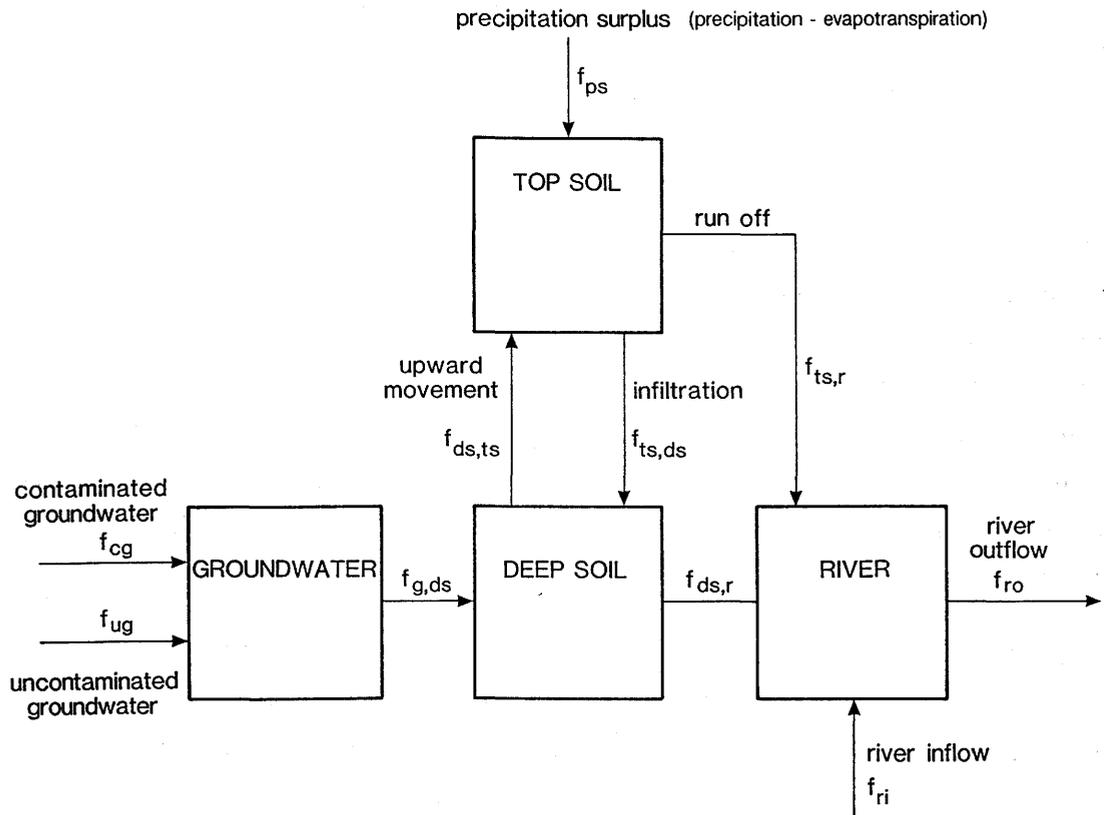


Figure 1: System of compartments for a two-layer soil model

and furthermore such soils are generally of minor importance agriculturally, often only being used as grassland. For our modelling purposes it is simply assumed that the groundwater is contaminated and its upward movement occurs in the region modelled (i.e. Laufenburg). The physical reasonableness of such a process at this generic site it not addressed.

## 2.2 Quantification of the transfer from groundwater to soil

The key parameter in this scenario is the extent of groundwater movement from the aquifer to the top soil. In Switzerland, the net water fluxes in the soil are mostly downwards because the total annual precipitation exceeds the yearly evapotranspiration. However, over shorter time periods transport upwards is possible when potential evapotranspiration exceeds precipitation. For a given time period the mass balance is:

$$F_d = (P-I) - ETP + \Delta W$$

in which  $F_d$  : water flux downwards [mm],  
 $P$  : precipitation [mm],  
 $I$  : interception [mm],  
 $ETP$  : evapotranspiration [mm],  
 $\Delta W$  : change in soil water content in top soil during a certain period [mm],

Important parameters with respect to the downward flux (or upward flux in cases where  $F_d$  is negative) are hydraulic conductivity and the water storage capacity of the soil. Soil characteristics which influence these parameters are particle and pore size distribution, soil depth, changes of soil properties with depth (layering) and the slope. The water storage capacity acts as buffer. The maximum available soil water is generally defined as the difference between soil moisture content at field capacity ( $pF = 1.8 - 2.5$ ) and at permanent wilting point ( $pF = 4.2$ ) (Scheffer and Schachtschabel, 1989).

As an example, the monthly water balances for the summer period (April to October 1971) and the water balance for the winter period (November 1971 to March 1972), given by Richard and Germann (1978) for a field in Möhlin (northern Switzerland) are shown in Table 1. The top soil depth is considered to be 40 cm for this Möhlin

evaluation (in the assessment model calculations 25 cm is assumed). The water balance in the deep soil is not given in this table. For a more detailed description of the experiment the reader is referred to the original literature.

Table 1: Water balance for a field in Möhlin in the summer period (monthly values) and the winter period in 1971 to 1972 (after Richard and Germann, 1978).

Month	P [mm]	I [mm]	ETP [mm]	$\Delta W$ [mm]	$F_d$ [mm]
April	24.1	1.5	28.1	-2.5	-8.0
May	47.9	7.7	36.8	-19.4	-16.0
June	135.1	18.4	131.6	5.3	-9.6
July	95.7	3.0	131.4	22.3	-16.4
August	129.7	7.3	105.7	-18.4	-1.7
September	46.8	6.3	42.6	5.1	3.0
October	30.4	2.4	45.4	9.2	-8.2
Summer total	509.7	46.6	521.6	1.6	-57.0
Winter total	211.9	13.0	70.0	-29.0	99.9

The experimental data in Table 1 shows that the downward flux during the summer months (with the exception of September) is negative and hence some upward movement occurs during this period. The summer total amounts to  $\sim 60$  mm. Thus during short periods of the year, groundwater could reach the top soil even when the net annual flow is downwards assuming the capillary fringe extends to the soil surface.

From this example the extent of upward movement can be evaluated and hence provide a reasonable basis for determining upward and downward fluxes to be considered in the model. Based on this study we assume that the upward flux of water from deep soil to top soil can range between 25 mm (minimum flux) and 100 mm (maximum flux) per year.

### 2.3 Evaluation of compartment volumes and water fluxes

The physical compartments and water fluxes in the base case calculations are based on the Laufenburg region in northern Switzerland. This region is situated on a broad gravel terrace along the south bank of the Rhine river. As mentioned above, this system is essentially the same as that used to assess the radiological impact of an underground

repository (NAGRA, 1985), and a general description already exists (Jiskra, 1985). The differences arising from the consideration of two soil layers rather than one are given below.

### 2.3.1 Compartment volumes

The surface area of the Laufenburg model region is 337.5 ha. The depth of the top soil corresponds to the rooting zone layer and is assumed to be 25 cm, whilst the deep soil compartment is assumed to be 1 m thick. It is assumed that both soil compartments contain 10% air, 30% water and 60% solid, by volume. The mass density of the minerals is taken as  $2650 \text{ kg m}^{-3}$ . The volume of the groundwater and river compartments are as evaluated by Jiskra (1985).

### 2.3.2 Water fluxes

The flow of the groundwater through the deep soil into the river was estimated from hydrological maps by Jiskra (1985). As discussed above, the flow from the deep soil to the top soil is assumed to range from 25 mm to 100 mm per year.

The annual precipitation is assumed to be 1000 mm in northern Switzerland and the evapotranspiration to be 500 mm (Jiskra, 1985). More detailed precipitation data, recorded in northern of Switzerland (Jaeggli and Frei, 1978), shows that about 40% of the rain falls during the winter period and 60% during the summer period. Evapotranspiration is higher during the summer (85% of the precipitation) and lower in the winter (10 to 60% of the precipitation, depending on agricultural practice). It should be noted, however, that high evapotranspiration will often cause a lowering of the water table with the possibility that the capillary fringe will not reach the surface. In this case the upward flux through the fringe will cease. For conservatism it is simply assumed that the capillary fringe remains in contact with the surface. It has also been observed that, during the vegetation period (4–5 months), only 5% of the measured rainfall passes through the soil whilst, in the winter period, about 85% percolates through the arable soil or grassland.

Taking these observations into account, for a rainfall regime with 500 mm precipitation surplus (i.e. total precipitation – evapotranspiration), it appears reasonable to consider that small upward movements are associated with high infiltration rates from top soil

to deep soil. For the base case scenario, we therefore consider a precipitation surplus of  $500 \text{ mm a}^{-1}$ , an infiltration of  $400 \text{ mm a}^{-1}$  and a capillary rise of  $25 \text{ mm a}^{-1}$ . A run-off of  $125 \text{ mm a}^{-1}$  flows directly into the river. The compartment volumes and the flows corresponding to this scenario (see also Figure 1) are summarised in Table 2. The data for two other cases are also given here but are discussed later in the report (see sections 4.3.4 and 5.1).

Table 2: Compartment volumes and waterflows for a two-layer soil model for various scenarios.

	Base Case	Site variation	Project Gewähr (1985)
	Laufenburg	Hellikon	Laufenburg
Compartment volumes [ $\text{m}^3$ ]			
Groundwater, $V_G$	$6.50 \times 10^6$	$2.63 \times 10^5$	$6.50 \times 10^6$
Deep soil, $V_{DS}$	$3.37 \times 10^6$	$2.78 \times 10^5$	$3.37 \times 10^6$
Top soil, $V_{TS}$	$8.44 \times 10^5$	$6.87 \times 10^4$	$8.44 \times 10^5$
River, $V_R$	$1.30 \times 10^6$	$1.30 \times 10^6$	$1.30 \times 10^6$
Waterflows [ $\text{m}^3\text{a}^{-1}$ ]			
contaminated groundwater, $f_{cg}$	$5.50 \times 10^6$	$2.63 \times 10^5$	$3.30 \times 10^6$
uncontaminated groundwater, $f_{ug}$	–	–	$2.20 \times 10^6$
groundwater to deep soil, $f_{g,ds}$	$5.50 \times 10^6$	$2.63 \times 10^5$	$5.50 \times 10^6$
upward movement, $f_{ds,ts}$	$8.45 \times 10^4$	$6.88 \times 10^3$	$8.45 \times 10^4$
precipitation surplus, $f_{ps}$	$1.69 \times 10^6$	$1.38 \times 10^5$	$1.69 \times 10^6$
run off, $f_{ts,r}$	$4.25 \times 10^5$	$3.49 \times 10^4$	$1.10 \times 10^6$
infiltration, $f_{ts,ds}$	$1.35 \times 10^6$	$1.10 \times 10^5$	$6.75 \times 10^5$
deep soil to river, $f_{ds,r}$	$6.77 \times 10^6$	$3.66 \times 10^5$	$6.09 \times 10^6$
river inflow, $f_{ri}$	$3.23 \times 10^{10}$	$3.23 \times 10^{10}$	$3.23 \times 10^{10}$
river outflow, $f_{ro}$	$3.23 \times 10^{10}$	$3.23 \times 10^{10}$	$3.23 \times 10^{10}$

### 3 Solid-liquid distribution coefficients and soil-to-plant transfer factors for $^{129}\text{I}$ and $^{237}\text{Np}$

The contamination of food products (root and leaf vegetables, cereals and pasture) will contribute to the dose to man. In the present model these are attached to the top soil compartment with which equilibrium is assumed. The key parameter for modelling this transfer from the soil to the plant is via the soil-to-plant transfer factor. However, this parameter depends on the solid-liquid distribution coefficient for a given radionuclide. An overview of the soil-plant relationship (after Van Loon, 1986) is schematically given in Figure 2.

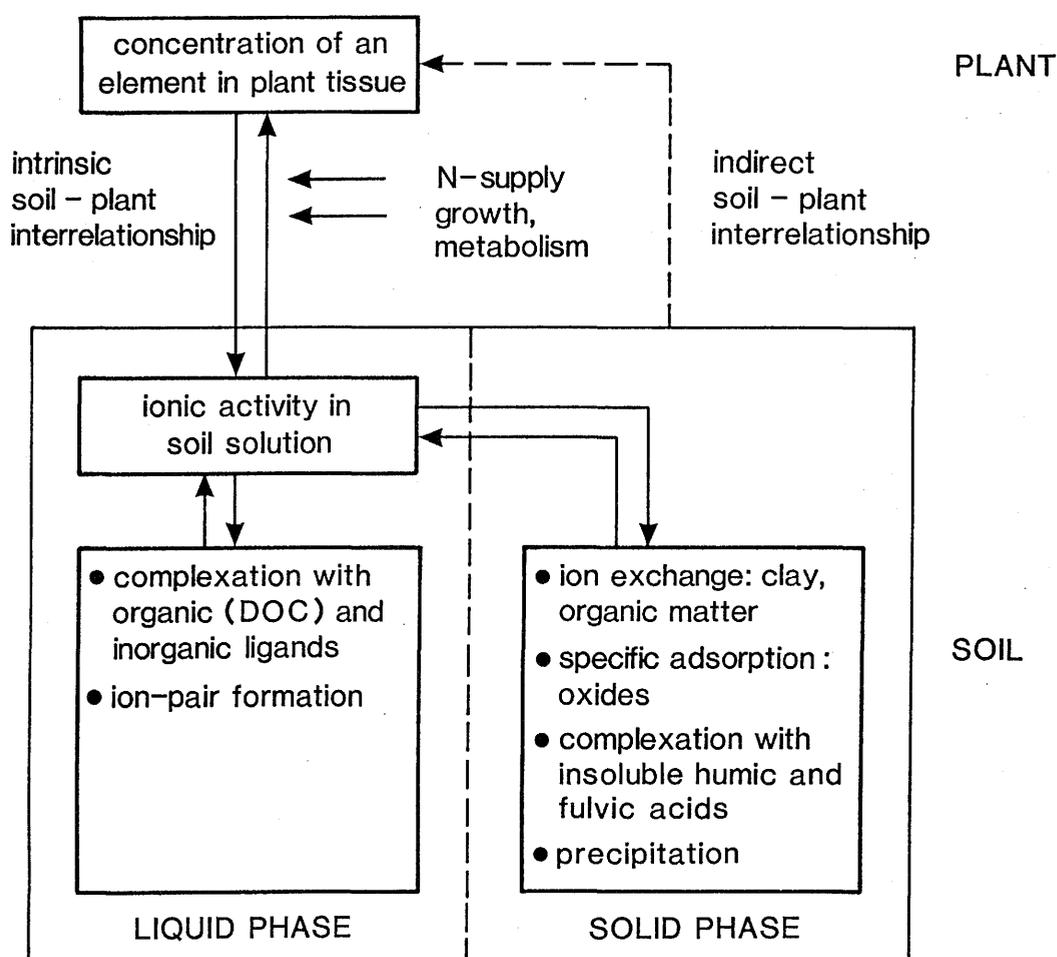


Figure 2: Schematic overview of the soil-plant relationship (taken from Van Loon, 1986)

The distribution of an element between the liquid phase and the solid phase is often represented by a distribution coefficient,  $K_d$ , expressed in  $\text{m}^3 \text{kg}^{-1}$ . The  $K_d$  value is a function of several interrelated parameters such as the chemical speciation, the nuclide concentration, soil type, redox conditions, sorption processes, and many others. The ion activity of an element in the soil solution is affected by both solid and liquid phases.

For some elements it has been shown that there is a direct relationship between the ion activity of an element in the soil solution and the resultant concentration in the plant (i.e. the intrinsic soil-plant interrelationship). Linear relationships have, for example, been observed for the uptake of technetium by spinach plants from hydroponic cultures (Van Loon, 1986). Similarly, there is indication that toxicity or bioavailability is more directly related to the free ion concentration (i.e. non complexed) rather than the total concentration of the element in the soil. However, it is recognised that the situation can be considerably more complex. For this reason this apparently more simple intrinsic interrelationship has not been adopted for modelling purposes and the transfer factor remains the most commonly used parameter to describe this process. However, by applying an inverse relation between the solid-liquid distribution coefficient and the soil-to-plant transfer factor a more realistic situation exists. It should be noticed that the soil-to-plant transfer factor is defined, and in most cases also measured, as the ratio of the concentration of an element in the plant and the soil (i.e. soil + soil solution). Thus, for a low  $K_d$  value (high soil solution concentration) a high transfer factor should be used and *vica versa*. Such a relationship has been proposed by various authors (Baes III, 1982, Sheppard, 1985 and Sheppard, 1986) and has also been experimentally observed (Sheppard et al. 1983). Applying such a relationship clearly reduces the overall uncertainty of the radiation dose to man. A brief overview of  $K_d$  and transfer factor values is given for both radionuclides and an inverse relationship between these factors is applied in the uncertainty calculations.

## 3.1 Data for Iodine

### 3.1.1 $K_d$ values

A literature survey has yielded a range of  $K_d$  values for iodine which are summarised in Figure 3. The data from Kocher (1982), Coughtrey et al. (1985), Jiskra (1985), AECL (1985) and Liu and von Gunten (1988) are themselves based on reviews. The data from Fleming (1980), which originate from Whitehead (1973), are experimentally measured.

The presented data cover only solid-liquid distribution coefficients for soil systems. However, no distinction is made for different soil types (e.g. sandy, loam, clay or organic). From the ranges reported, minimum and maximum  $K_d$  values are selected whilst a value of  $0.01 \text{ m}^3 \text{ kg}^{-1}$  is taken as a best estimate value.

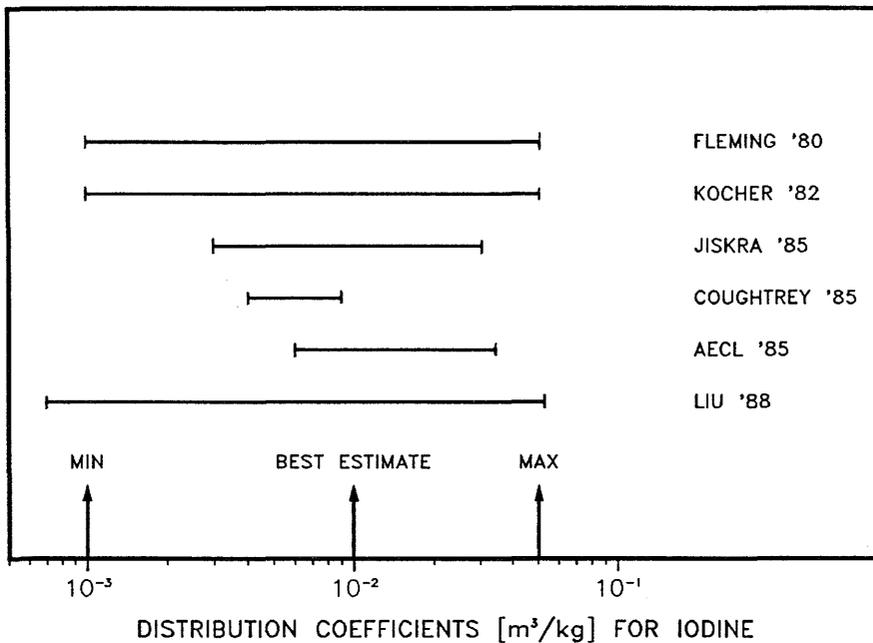


Figure 3: Literature data for solid-liquid distribution coefficients for iodine

### 3.1.2 Soil-to-plant transfer factors

Soil-to-plant transfer factors for iodine were reviewed by Grogan (1985). The best estimate values from this review are presented in Table 3.

The compilations from Ng et al. (1982) and Coughtrey et al. (1983) provide data for soil-to-plant transfer factors for iodine. The transfer factors for the various crops are consistent between the two reviews and range over 2 to 3 orders of magnitude. Both data sets are used in order to obtain minimum and maximum values of the iodine transfer factor. Table 3 summarises the selected values.

The minimum and maximum transfer factors reported for iodine cover the values of the reviewed Swedish biosphere data base from Bergström et al. (1986).

Table 3:  $K_d$  and soil-to-plant transfer factors (TF) for iodine

	Best estimate	Maximum $K_d$ Minimum TF	Minimum $K_d$ Maximum TF
$K_d$ [ $\text{m}^3 \text{kg}^{-1}$ ]	$1.0 \times 10^{-2}$	$5.0 \times 10^{-2}$	$1.0 \times 10^{-3}$
TF: root crops/soil	$5.6 \times 10^{-3}$	$1.4 \times 10^{-3}$ <sup>a)</sup>	$2.0 \times 10^{-1}$ <sup>b)</sup>
TF: leaf crops/soil	$1.9 \times 10^{-2}$	$1.0 \times 10^{-3}$ <sup>b)</sup>	$2.0 \times 10^{-1}$ <sup>b)</sup>
TF: cereals/soil	$3.6 \times 10^{-1}$	$1.6 \times 10^{-2}$ <sup>a)</sup>	$9.8 \times 10^{-1}$ <sup>a)</sup>
TF: pasture/soil	$1.0 \times 10^{-1}$	$8.8 \times 10^{-3}$ <sup>a)</sup>	$2.4 \times 10^{-1}$ <sup>a)</sup>

a) Ng et al. (1982).

b) Coughtrey et al. (1983).

## 3.2 Data for Neptunium

### 3.2.1 $K_d$ values

A literature survey of distribution coefficients for neptunium is given in Figure 4. Experimental data are from Sheppard et al. (1979), Nishita et al. (1981) and Henrion et al. (1985). The remaining references are review data. The highest values given (Henrion et al. 1985) are measurements on a marine clay sediment (under strongly reducing conditions) and are only relevant for clay rich soils under anoxic conditions. For this reason, the maximum  $K_d$  value considered is  $1 \text{ m}^3 \text{kg}^{-1}$ . The lowest values refer to sandy soils. Because soils in northern Switzerland always contain a certain amount of clay, silt and organic matter a range from 0.001 to  $1 \text{ m}^3 \text{kg}^{-1}$  was chosen.

### 3.2.2 Soil-to-plant transfer factors

The transfer factors for neptunium were reviewed by Grogan (1985) and Grogan and van Dorp (1986). The former reference present best estimate values whilst the latter also included ranges. These data sets are accepted here. As for iodine, Table 4 relates the  $K_d$  values with the transfer factors.

Table 4:  $K_d$  and soil-to-plant transfer factors (TF) for neptunium

	Best estimate	Maximum $K_d$ Minimum TF	Minimum $K_d$ Maximum TF
$K_d$ [ $\text{m}^3 \text{kg}^{-1}$ ]	$5.0 \times 10^{-2}$	$1.0 \times 10^0$	$1.0 \times 10^{-3}$
TF: root crops/soil	$6.0 \times 10^{-2}$	$3.0 \times 10^{-4}$	$3.0 \times 10^{-1}$
TF: leaf crops/soil	$2.7 \times 10^{-2}$	$4.5 \times 10^{-4}$	$1.3 \times 10^{-1}$
TF: cereals/soil	$1.7 \times 10^{-2}$	$4.5 \times 10^{-5}$	$2.5 \times 10^{-1}$
TF: pasture/soil	$9.4 \times 10^{-3}$	$6.3 \times 10^{-4}$	$1.4 \times 10^{-1}$

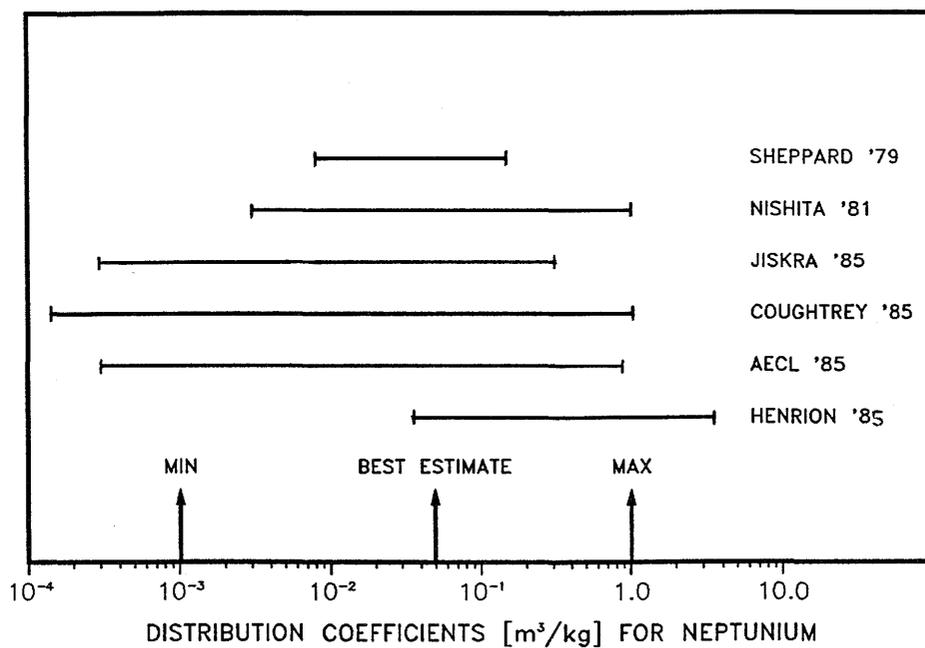


Figure 4: Literature data for solid-liquid distribution coefficients for neptunium

## 4 BIOMOVs scenario B6: Transport of contaminated groundwater to soil

The BIOMOVs B6 scenario deals with the upward movement of radionuclides from deep soil to surface soils, and hence crops and the atmosphere. The basis for this scenario is the same as for the groundwater model described in section 2.1 of this report. The data evaluated in sections 2.2 and 2.3 and chapter 3 are used for the base case calculations. A series of parameter variations are included in order to quantify uncertainty in this scenario. The results of the calculations and uncertainty are presented below.

### 4.1 Definition of the scenario B6

The following scenario definition was supplied to the BIOMOVs participants:

**There is a flux of 1 Bq a<sup>-1</sup> hectare<sup>-1</sup> of <sup>237</sup>Np and <sup>129</sup>I in groundwater to soil below the rooting zone in a homogeneous farming area where there is no irrigation. Calculate concentrations in rooting zone soil (Bq kg<sup>-1</sup> dry wt.), root crops (Bq kg<sup>-1</sup> fresh wt. prepared for human consumption) and the atmosphere (Bq m<sup>-3</sup> as inhaled) until a steady-state is reached.**

All other parameter values were defined based on the judgement of the group participants in BIOMOVs.

One of the primary objectives of the intercomparison study "BIOMOVs" is to test the accuracy of predictions of environmental assessment models for selected contaminants and exposure scenarios and to explain differences in the predictions due to model deficiencies, invalid assumptions and/or differences in selected input data (BIOMOVs, 1986). In order to facilitate the intercomparison, the most important processes with respect to this scenario are summarised in a simplified manner in Figure 5.

In accordance with the definition, the radionuclide input from the groundwater to the deep soil (via groundwater) is fixed at 1 Bq a<sup>-1</sup> hectare<sup>-1</sup>. The compartment volumes and the water flows into and between the compartments are already discussed and are given in Table 2 (base case: Laufenburg) and apply to the whole model region.

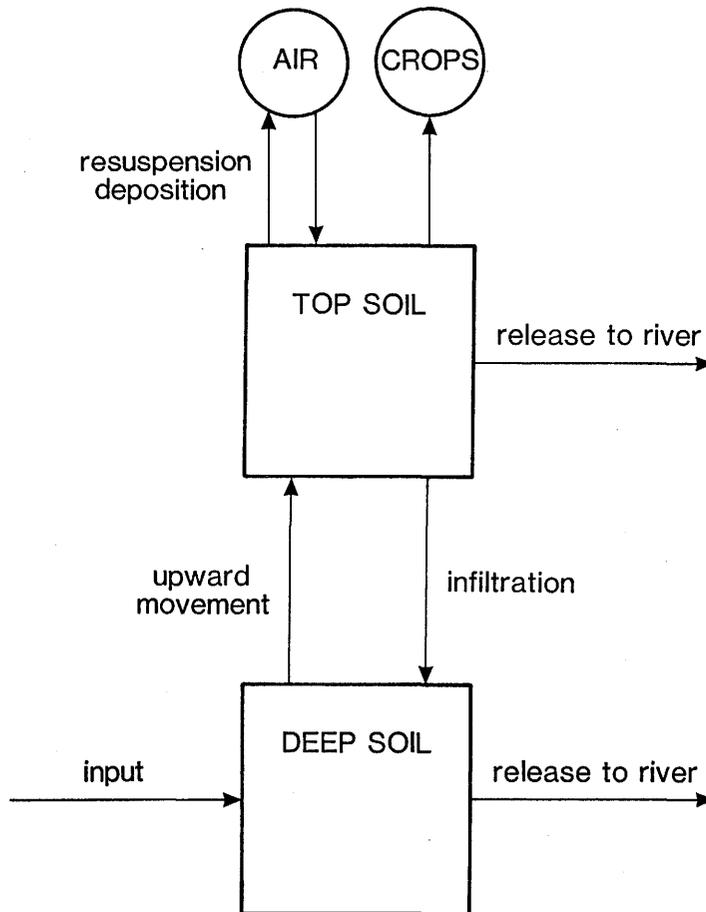


Figure 5: Water and nuclide fluxes for test scenario B6 model comparison

The radionuclide concentration in the deep and top soil compartments are calculated as a function of time until steady-state is reached. The solid-liquid distribution coefficients for both soil layers are the best estimate values given in Tables 3 and 4.

In this model, the foodcrops are attached to the top soil compartment and simple transfer factors are applied. For this reason, the concentrations in root vegetables, leaf vegetables, cereals and pasture are only calculated at steady-state. The soil-to-plant transfer factors used for these calculations are the best estimates given in Tables 3 and 4.

The dust load in the air is assumed to be  $50 \mu\text{g m}^{-3}$ . This value was also adopted by Grogan and van Dorp (1986) in the BIOMOVS B2 test scenario (BIOMOVS, 1989) and is representative of typical dust load concentrations. Deposition of potentially contaminated soil is not considered in the model. The air concentration is also only calculated at steady-state.

Tables 5 to 7 summarises the relevant water fluxes and parameters used in this calculation

with respect to the BIOMOVs scenario definition. The soil-to-plant transfer factors for leaf vegetables, cereals and pasture (not specified in the scenario description) are included for completeness.

Table 5: Water fluxes for the base case calculations of scenario B6.

Water flux	[m <sup>3</sup> a <sup>-1</sup> ha <sup>-1</sup> ]
Groundwater to deep soil (input)	16250
Deep soil to top soil (upward movement)	250
Top soil to deep soil (infiltration)	4000
Top soil to river (runoff)	1250
Deep soil to river (drainage)	20000

Table 6: Soil parameters used for the base case calculations of scenario B6.

Soil physical characteristics	Deep soil	Top soil
Depth of soil layer [m]	1.0	0.25
Porosity [-]	0.4	0.4
Bulk density [kg m <sup>-3</sup> ]	1590	1590
Water filled pore space [-]	0.3	0.3
Density of minerals [kg m <sup>-3</sup> ]	2650	2650

Table 7: Solid-liquid distribution coefficient and soil-to-plant transfer factors used for the base case calculations of scenario B6

Parameters	<sup>237</sup> Np	<sup>129</sup> I
Top soil K <sub>d</sub> [m <sup>3</sup> kg <sup>-1</sup> ]	5.0 x 10 <sup>-2</sup>	1.0 x 10 <sup>-2</sup>
Deep soil K <sub>d</sub> [m <sup>3</sup> kg <sup>-1</sup> ]	5.0 x 10 <sup>-2</sup>	1.0 x 10 <sup>-2</sup>
TF: root vegetables/soil	6.0 x 10 <sup>-2</sup>	5.6 x 10 <sup>-3</sup>
TF: leaf vegetables/soil	2.7 x 10 <sup>-2</sup>	1.9 x 10 <sup>-2</sup>
TF: cereals/soil	1.7 x 10 <sup>-2</sup>	3.6 x 10 <sup>-1</sup>
TF: pasture/soil	9.4 x 10 <sup>-3</sup>	1.0 x 10 <sup>-1</sup>

## 4.2 Results of base case calculations and discussion

The evolution of the nuclide concentrations in both soil layers as a function of time is illustrated in Figures 6 for neptunium and iodine. The radionuclide concentration increases in both soil layers smoothly as a function of time, until a steady-state is reached. The most striking feature in these figures is that the top soil concentrations are lower than the deep soil concentrations. This is because the amount of contaminated water reaching the top soil is only a fraction of that reaching the deep soil. This can be clearly seen from the data presented in Table 8 where the nuclide fluxes and transfer coefficients ( $K_{n,m}$ ) are shown.

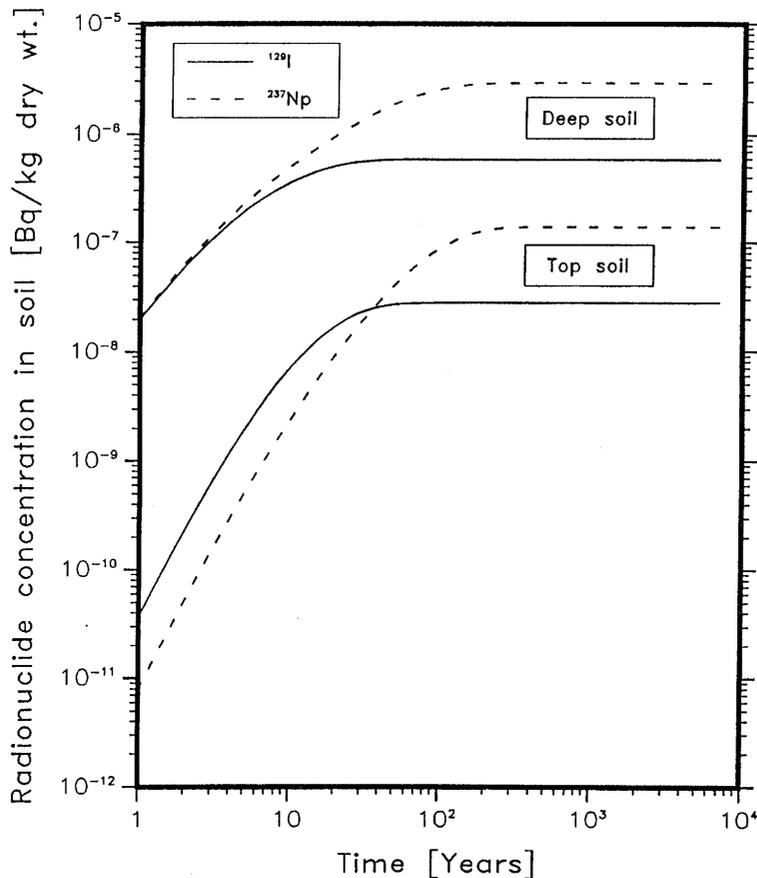


Figure 6: Soil concentrations as a function of time for  $^{237}\text{Np}$  and  $^{129}\text{I}$  in the base case scenario.

Table 8: Radionuclide fluxes and transfer coefficients for the base case calculations.

Transfer	Flux [Bq a <sup>-1</sup> ha <sup>-1</sup> ]	$K_{n,m}$ [a <sup>-1</sup> ]
Groundwater to deep soil	1.000	$8.5 \times 10^{-1}$
Deep soil to top soil	0.012	$2.7 \times 10^{-4}$
Top soil to deep soil	0.009	$1.7 \times 10^{-2}$
Top soil to river	0.003	$5.4 \times 10^{-3}$
Deep soil to river	0.997	$2.2 \times 10^{-2}$

The transfer coefficient,  $K_{n,m}$ , is the fraction of radionuclide that goes per time unit from compartment n to compartment m. In the case when no retardation occurs, i.e. the radionuclide is passively transported with the water,  $K_{n,m}$  is defined as:

$$K_{n,m} = \frac{F_{n,m}}{V_n} \quad [a^{-1}]$$

where:  $F_{n,m}$  = waterflux [m<sup>3</sup> a<sup>-1</sup>]  
 $V_n$  = volume of compartment n [m<sup>3</sup>]

In the case with retardation, i.e. the radionuclide is sorbed on the soil solids,  $K_{n,m}$  is defined as:

$$K_{n,m} = \frac{F_{n,m}}{(V_w + \rho_s K_d V_s)} \quad [a^{-1}]$$

where:  $V_w$  = volume of the soil water [m<sup>3</sup>]  
 $V_s$  = volume of the soil solid [m<sup>3</sup>]  
 $\rho_s$  = density of the soil solid [kg m<sup>-3</sup>]  
 $K_d$  = solid-liquid distribution coefficient [m<sup>3</sup> kg<sup>-1</sup>]

It should also be noted that the  $K_d$ 's assumed are the same for both soil layers. At early times after release has commenced, the top soil concentration is 3 orders of magnitude less than that in the deep soil. This difference decreases as a function of time and, at steady-state, the difference between the deep and top soil concentrations is a factor of 20. Since it is assumed in this model that the deep soil does not represent a direct pathway to man, the resultant contamination of crops growing on the top soil appears less hazardous than would be the case for a single soil compartment model. In the latter, the deep soil concentration would represent the total soil contamination. Clearly

now that the foodchain exposure pathways are less important, direct exposure pathways such as drinking water will assume greater significance.

Table 9 presents the results for the base case calculations for  $^{237}\text{Np}$  and  $^{129}\text{I}$ . The difference in soil concentration between these nuclides stems from the different  $K_d$  values used (see section 4.3.3).

Table 9: Steady-state results for the base case calculations.

Steady-state concentration	$^{237}\text{Np}$	$^{129}\text{I}$
Root zone soil [ $\text{Bq kg}^{-1}$ dry]	$1.4 \times 10^{-7}$	$2.8 \times 10^{-8}$
Deep soil [ $\text{Bq kg}^{-1}$ dry]	$2.9 \times 10^{-6}$	$5.9 \times 10^{-7}$
Root vegetables [ $\text{nBq kg}^{-1}$ ]	8.4	0.2
Leaf vegetables [ $\text{nBq kg}^{-1}$ ]	3.8	0.5
Cereals [ $\text{nBq kg}^{-1}$ ]	2.4	10.1
Pasture [ $\text{nBq kg}^{-1}$ dry]	1.3	2.8
Air [ $\text{nBq m}^{-3}$ ]	7.0	1.4
Time to reach steady-state [a]	360	100

### 4.3 Parameter variations

A number of parameter variations have been considered, with the aim of gaining more understanding about the behaviour of the system and providing an estimate of uncertainty in relation to the radiological impact. However, since only a limited number of key factors are taken into account a prediction of the real uncertainty is not obtained. Parameter variations with respect to climate (infiltration flux), groundwater flow, sorption ( $K_d$  values) and compartment volumes are described and their results are presented below.

#### 4.3.1 Effect of reduced infiltration

In this variation it is simply assumed that the precipitation surplus onto the top soil is decreased from  $500 \text{ mm a}^{-1}$  to  $250 \text{ mm a}^{-1}$ . Such a decrease could correspond

to a somewhat warmer climate. The total annual precipitation is still assumed to be 1000 mm and, therefore, the annual evapotranspiration is increased to 750 mm. The low rainfall regime is assumed to result in an increase in the capillary rise (100 mm instead of 25 mm) and a lower infiltration rate from top to deep soil (50 mm instead of 400 mm).

Figure 7 illustrates the effect on the neptunium top soil concentration assuming either  $500 \text{ mm a}^{-1}$  (i.e. base case) or  $250 \text{ mm a}^{-1}$  precipitation surplus. Clearly, a low rainfall regime results in the highest top soil concentration. In this variation, different factors are contributing to the increased soil concentration. The most important factor is the higher nuclide flux upwards (due to the increased upward flux of contaminated groundwater). The lower precipitation surplus, combined with the lower infiltration rate, both lead to a decreased dilution and hence, increased top soil contamination. The overall effect is a seven fold increase in top soil concentration compared with the base case calculations. The behaviour of iodine with regard to this and following parameter variations is analogous and is not discussed here.

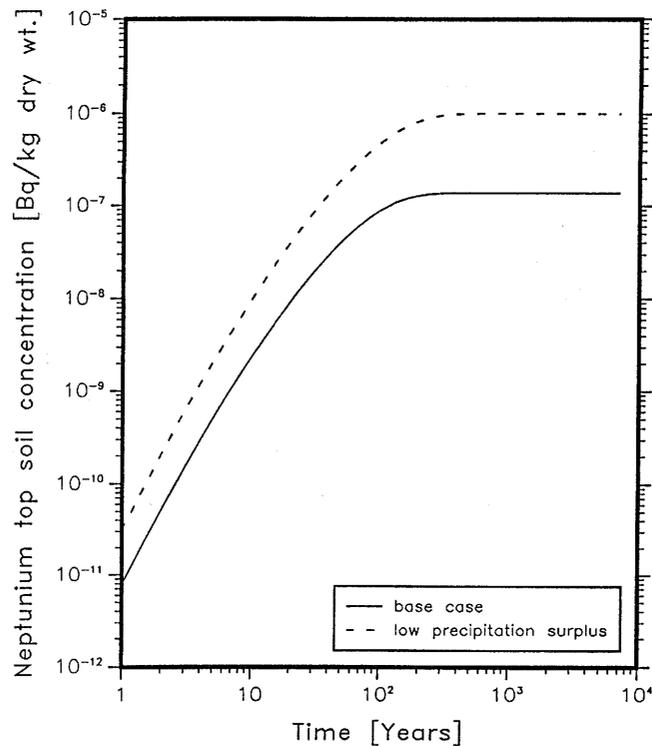


Figure 7: Effect of varying precipitation on the neptunium top soil concentration

### 4.3.2 Effect of decreased groundwater flow

In this variation it is assumed that the groundwater flux is 10 times less than in the base case. The concentration of radionuclides in the groundwater is then 10 times greater since the input radionuclide flux of  $1 \text{ Bq a}^{-1} \text{ hectare}^{-1}$  is maintained. This variation is considered since it is known that the hydraulic conductivity of the near-surface sediments may vary by at least one order of magnitude (Hydrogeologische Karte der Schweiz, 1972). This calculation therefore represents release into a deep aquifer whose hydraulic conductivity is 10 times smaller than in the base case. The results are shown in Figure 8.

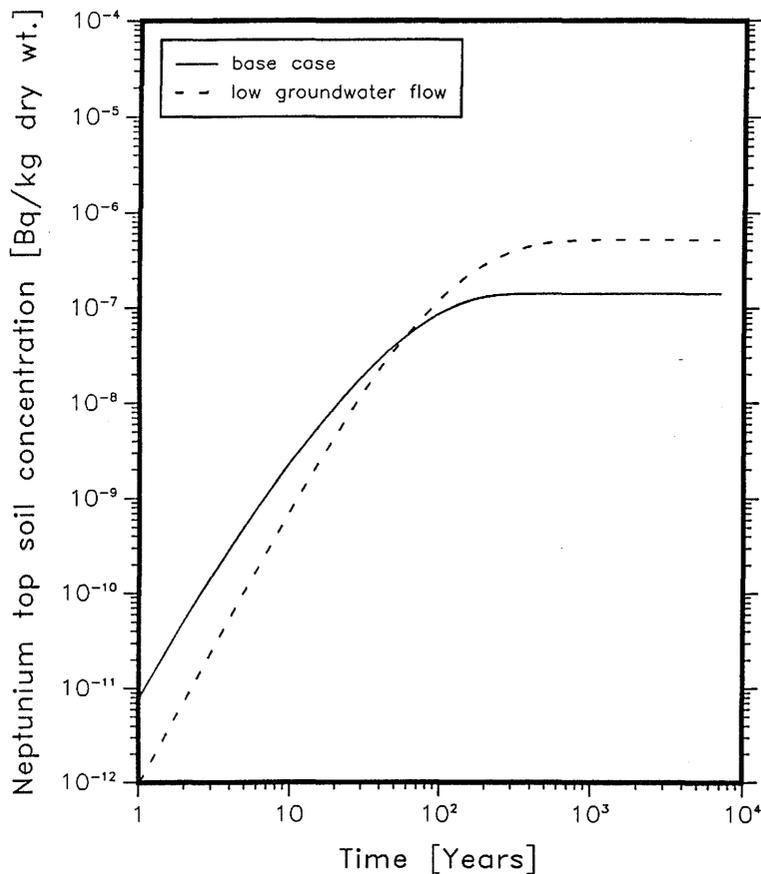


Figure 8: Effect of a reduced groundwater flow on the neptunium soil concentrations

Figure 8 shows the time dependent root zone soil concentrations for neptunium for this variation. Even though the groundwater concentration is enhanced by one order of magnitude, the soil concentration only increases by a factor of about 4. This is because

two processes are operating simultaneously. Firstly, there is increased contamination of the deep and top soil due to the higher groundwater radionuclide concentrations. However, at the same time, the release of radioactivity from the deep and top soil to the river also increases compared with the base case. This is illustrated in Table 10 where the neptunium fluxes and transfer coefficients (i.e. rate constants) of this scenario are compared with the base case. The combination of these interrelated processes results in an overall increase of the nuclide concentration in both soil layers by a factor of 4. The time to reach steady-state is also increased by the same factor.

Table 10: Np fluxes and transfer coefficients ( $K_{n,m}$ )

Transfer	Base case		Low groundwater flow	
	Flux [Bq a <sup>-1</sup> ha <sup>-1</sup> ]	$K_{n,m}$ [a <sup>-1</sup> ]	Flux [Bq a <sup>-1</sup> ha <sup>-1</sup> ]	$K_{n,m}$ [a <sup>-1</sup> ]
Groundwater to deep soil	1.000	$8.5 \times 10^{-1}$	1.000	$8.5 \times 10^{-2}$
Deep soil to top soil	0.012	$2.7 \times 10^{-4}$	0.046	$2.7 \times 10^{-4}$
Top soil to deep soil	0.009	$1.7 \times 10^{-2}$	0.035	$1.7 \times 10^{-2}$
Top soil to river	0.003	$5.4 \times 10^{-3}$	0.011	$5.4 \times 10^{-3}$
Deep soil to river	0.997	$2.2 \times 10^{-2}$	0.989	$5.8 \times 10^{-3}$

### 4.3.3 Effect of variation in $K_d$ values

A number of variations of  $K_d$  have been carried out for the Laufenburg base case scenario. In a first series, the best estimate  $K_d$  value is used in the deep soil but the minimum and maximum values, as given in Tables 3 and 4, are used for the top soil. Figure 9 shows the effect of this variation for neptunium. These results clearly demonstrate two effects: Firstly, the linear relationship between the top soil concentration and the  $K_d$  value used. A high  $K_d$  causes a high top soil concentration whilst the reverse effect is observed for a low  $K_d$  value. The higher  $K_d$  dictates a stronger association with the soil solid which ultimately leads to a higher level of soil contamination. Secondly, the time to reach steady-state also depends upon the  $K_d$ -value. This is because the radionuclide accumulation in the soil proceeds more slowly when higher sorption is specified.

Table 11 presents the transfer coefficients ( $K_{n,m}$ ) of the neptunium fluxes between the relevant compartments (see Figure 5) for the three different cases considered. This table shows that only the neptunium fluxes from top soil to deep soil and to the river

are varied. In the case a low top soil  $K_d$  is considered (i.e. low retardation), high radionuclide fluxes out of this compartment are given. The reverse effect is seen if a high top soil  $K_d$  is taken.

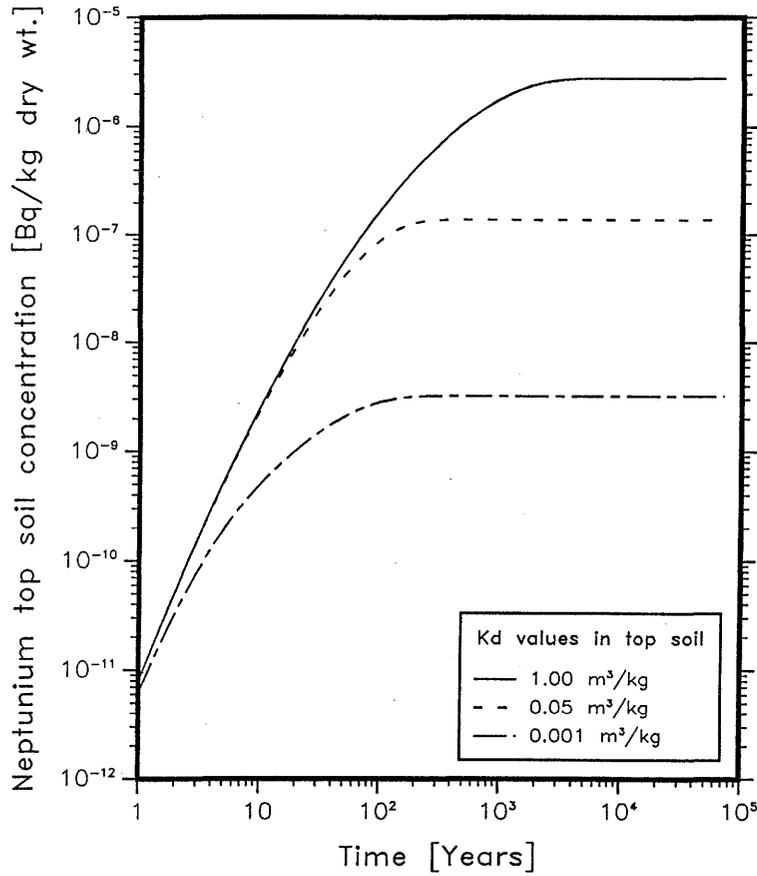


Figure 9: Effect of  $K_d$  on neptunium root zone soil concentrations

Table 11: Transfer coefficients for neptunium as function of top soil  $K_d$

Transfer	Minimum $K_d$	Base case $K_d$	Maximum $K_d$
	$K_{n,m}$ [a <sup>-1</sup> ]	$K_{n,m}$ [a <sup>-1</sup> ]	$K_{n,m}$ [a <sup>-1</sup> ]
Groundwater to deep soil	$8.5 \times 10^{-1}$	$8.5 \times 10^{-1}$	$8.5 \times 10^{-1}$
Deep soil to top soil	$2.7 \times 10^{-4}$	$2.7 \times 10^{-4}$	$2.7 \times 10^{-4}$
Top soil to deep soil	$7.4 \times 10^{-1}$	$1.7 \times 10^{-2}$	$8.6 \times 10^{-4}$
Top soil to river	$2.3 \times 10^{-1}$	$5.4 \times 10^{-3}$	$2.7 \times 10^{-4}$
Deep soil to river	$2.2 \times 10^{-2}$	$2.2 \times 10^{-2}$	$2.2 \times 10^{-2}$

In a second series of variations, the  $K_d$  values of the deep soil are varied but the top soil  $K_d$  is held constant at the best estimate value. Such a situation can occur, for example, for elements that exhibit redox-sensitive sorption behaviour. Reducing conditions in the deep soil layer may lead to higher sorption compared with oxidizing conditions in top soil (such a behaviour is expected for neptunium). The opposite might be considered for nuclides which may strongly interact with organic matter. In the case for iodine the retention in the top soil is expected to be higher than in the deep soil (less organic matter) (Fleming, 1980). The results of this variation are shown in Figure 10.

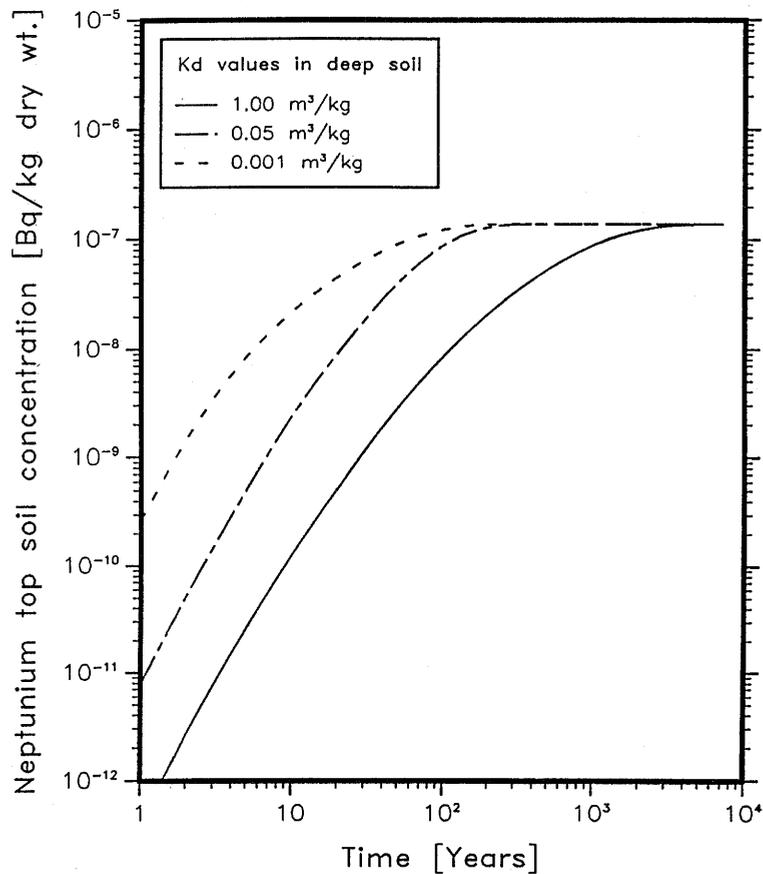


Figure 10: Effect of variation in deep soil  $K_d$  on neptunium root zone soil concentrations (top soil  $K_d$  remains constant)

Clearly, this figure shows that, at steady-state, the nuclide concentrations in the top soil in all cases are the same. In other words the sorption behaviour of a nuclide in the deep

soil does not affect the resultant concentration in the top soil layer under the conditions defined in this scenario. However, the time taken to reach steady-state concentrations is strongly influenced by the deep soil  $K_d$  used. The reason for this is that the time required to reach steady-state in the top soil is related with the time taken to reach steady-state in the deep soil. Table 12 and 13 summarises the results of these  $K_d$  variations for neptunium and iodine, respectively. The concentrations in the root zone soil, crops and air are also given. Since the  $K_d$  - transfer factor values are inversely related for the model calculations, the variation in the food crop concentration is much smaller than for the root zone soil and air concentrations. The latter are directly related to the  $K_d$  values used for the top soil layer.

Table 12: Effect of top soil  $K_d$  on the  $^{237}\text{Np}$  concentration in soil, crops and air

Top soil $K_d$ [ $\text{m}^3 \text{kg}^{-1}$ ]	Time to reach Steady-state [a]	Top Soil Concentration [ $\text{Bq kg}^{-1}$ ]	Crop Concentration Root Leaf Grain Pasture [ $\text{nBq kg}^{-1}$ ]				Air Conc. [ $\text{nBq m}^{-3}$ ]
0.001	14	$3.2 \times 10^{-9}$	1.0	0.4	0.8	0.5	0.2
0.05	360	$1.4 \times 10^{-7}$	8.4	3.8	2.4	1.3	7.0
1.0	7200	$2.8 \times 10^{-6}$	0.8	1.3	0.1	1.8	140

Table 13: Effect of top soil  $K_d$  on the  $^{129}\text{I}$  concentration in soil, crops and air

Top soil $K_d$ [ $\text{m}^3 \text{kg}^{-1}$ ]	Time to reach Steady-state [a]	Top Soil Concentration [ $\text{Bq kg}^{-1}$ ]	Crop Concentration Root Leaf Grain Pasture [ $\text{nBq kg}^{-1}$ ]				Air Conc. [ $\text{nBq m}^{-3}$ ]
0.001	14	$3.2 \times 10^{-9}$	0.6	0.6	3.1	0.8	0.2
0.01	100	$2.8 \times 10^{-8}$	0.2	0.5	10.1	2.8	1.4
0.05	360	$1.4 \times 10^{-7}$	0.2	0.1	2.2	1.2	7.0

#### 4.3.4 Effect of decreasing compartment volumes (Hellikon Region)

Another region in northern Switzerland (Hellikon) has been modelled in the same way as the base case. This is a smaller area with lower groundwater fluxes, so that the

compartment volumes and fluxes between them are also smaller. The compartmental scheme for this model region is identical to the Laufenburg model region and is shown in Figure 2. The compartment volumes and water fluxes are summarised in Table 2. Analogous to the base case, a  $500 \text{ mm a}^{-1}$  precipitation surplus and a  $25 \text{ mm a}^{-1}$  upward movement is assumed.

Figure 11 shows the neptunium and iodine concentration in both soil layers as a function of time assuming the groundwater enters the Hellikon region. The effect is very similar to decreasing the groundwater flow but less pronounced. Similarly there is a higher nuclide flux from groundwater to deep and top soil which causes higher soil contamination ( $\sim 50\%$  higher compared to the Laufenburg base case results). In comparison with the base case the smaller groundwater flow into the smaller soil compartments leads to higher nuclide transfer fluxes since the radionuclide source of  $1 \text{ Bq a}^{-1} \text{ hectare}^{-1}$  is maintained.

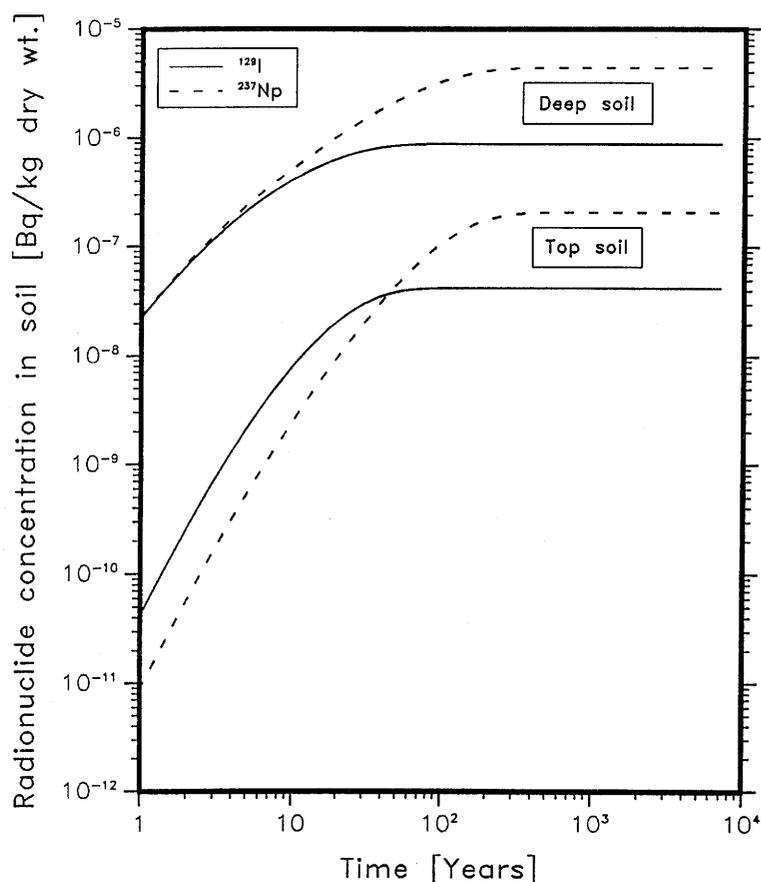


Figure 11: Soil concentration for iodine and neptunium in the Hellikon model region.

## 4.4 Uncertainty

Within the BIOMOVS B6 scenario, it is requested that the calculations should be represented as a best estimate values with their associated uncertainties. In this section uncertainty is discussed with emphasis being placed on uncertainties caused by the variability in key parameter values.

For the parameter perturbations considered, the variation in most results is either directly proportional or less than proportional to the extent of variation in the input parameter value. Of the parameter variations studied, the uncertainty caused by the  $K_d$  is the most important. Variations in the waterflow, infiltration and compartment sizes are either small or can be reduced by site specific informations. However, variations in  $K_d$  may be large due to problems in experimental measurements of values which are representative for the field.

Thus the uncertainty caused by the range in  $K_d$  is considered to be the single most important parameter. However, due to the assumed inverse relation between sorption ( $K_d$ ) and the soil-to-plant transfer factor, the uncertainty in the crop concentrations is smaller than the uncertainty in the soil concentrations. More investigations are necessary to supply data supporting this and to allow a better understanding of this inverse relationship.

No estimate has been made of the uncertainties in the dust load values for the air or the assumption that this dust is contaminated to the same level as the surface soil. Therefore, the variation of the calculated air concentrations of radionuclides is directly proportional to the top soil concentrations simply reflecting the uncertainty in the  $K_d$  value.

Table 14 summarises the concentration of neptunium and iodine in the root zone soil, in the root crops and in the atmosphere at steady-state as requested by the BIOMOVS B6 scenario definition. Best estimate values together with the minimum and maximum values are presented.

Table 14: Steady-state concentrations of  $^{237}\text{Np}$  and  $^{129}\text{I}$  in root zone soil, root crops and in the air. Best estimate values and uncertainty estimates

Nuclide		Root zone soil (Bq kg <sup>-1</sup> dry)	Root crops (Bq kg <sup>-1</sup> fresh)	Air (Bq m <sup>-3</sup> )
$^{237}\text{Np}$	Best estimate	$1.4 \times 10^{-7}$	$8.4 \times 10^{-9}$	$7.0 \times 10^{-15}$
	Minimum	$3.2 \times 10^{-9}$	$8.0 \times 10^{-10}$	$2.0 \times 10^{-16}$
	Maximum	$2.8 \times 10^{-6}$	$8.4 \times 10^{-9}$	$1.4 \times 10^{-13}$
$^{129}\text{I}$	Best estimate	$2.8 \times 10^{-8}$	$2.0 \times 10^{-10}$	$1.4 \times 10^{-15}$
	Minimum	$3.2 \times 10^{-9}$	$2.0 \times 10^{-10}$	$2.0 \times 10^{-16}$
	Maximum	$1.4 \times 10^{-7}$	$6.0 \times 10^{-10}$	$7.0 \times 10^{-15}$

## 5 Radiation dose calculations: a comparison with a reference case (Project Gewähr)

The main objective of Project Gewähr (NAGRA 1985) was to demonstrate that nuclear waste can be safely and permanently disposed in deep underground rock formations.

Radioactivity can return to the biosphere from a repository via dissolution in groundwater which might migrate through the geosphere and eventually reach the earth's surface. If such a process occurs, the soil would retard the movement of any nuclide released from the groundwater. This retardation must be quantified to predict the final dose to man. The biosphere model which was used to make these calculations is described in the following section. For the BIOMOVS intercomparison exercise, however, the calculations are not carried through as far as dose or risk, but stop at concentrations in root zone soil and foodcrops.

In this section, the approach with two soil compartments described in chapter 2 is applied to the situation as presented in Project Gewähr (NAGRA, 1985). Thus doses to man are calculated for the neptunium decay chain ( $^{237}\text{Np}$  -  $^{233}\text{U}$  -  $^{229}\text{Th}$ ) and the results are compared with those obtained in the Project Gewähr study.

### 5.1 Compartments and water flow data

In Project Gewähr 1985, a biosphere compartment model with one soil layer was used. The flow diagram for this reference case is shown schematically in Figure 12 together with the attached foodchains. This Laufenburg model region is basically the same as the one used throughout this report (see chapter 2), however, an additional flow of uncontaminated water is accounted for in this scenario.

The system of physical compartments for a two-layer soil model is applicable to the model given in Figure 2. The compartment volumes and flows are included in Table 2. The deep soil is assumed to be 1 m thick and has the same physical characteristics as the top soil. As with the previous base case calculations the upward flux from deep soil to top soil is assumed to be 25 mm per year.

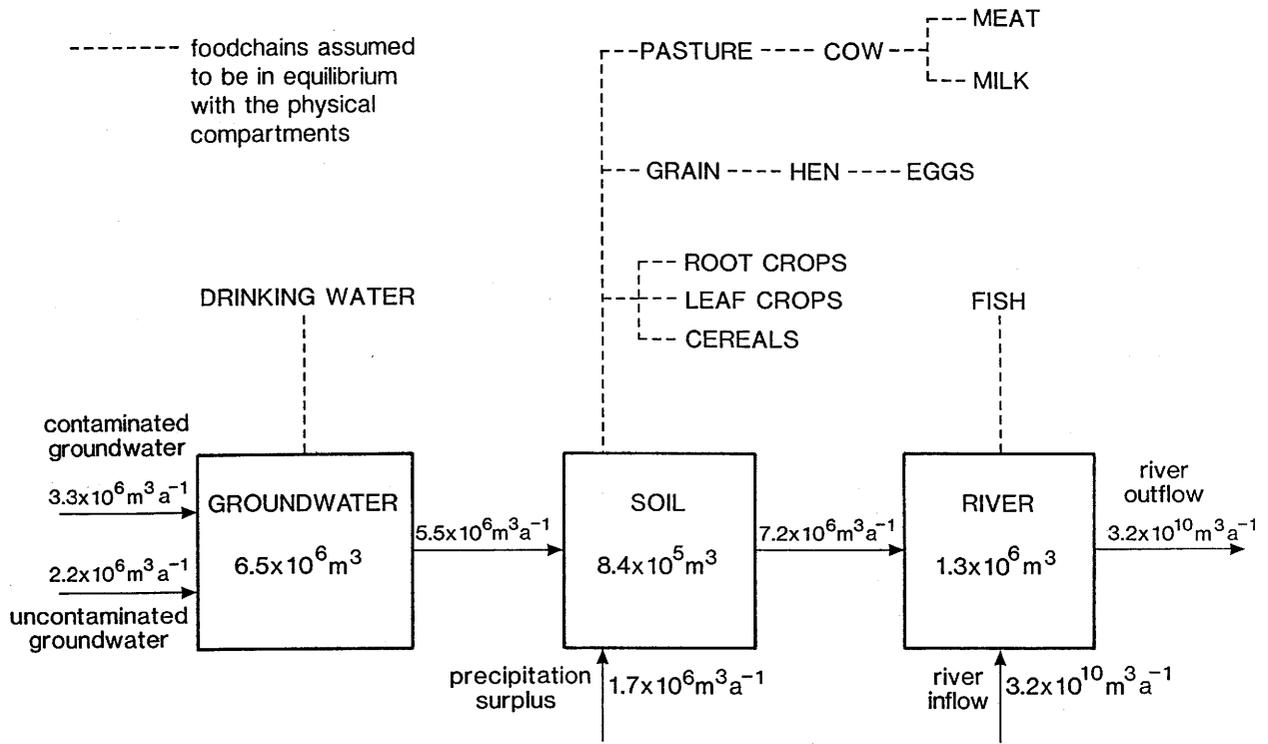


Figure 12: Compartements and foodchain pathways for the Laufenburg model region in a one-layer soil model (NAGRA 1985)

## 5.2 Results and discussion

### 5.2.1 Effect on root zone soil concentration

The radionuclide concentrations in the root zone soil to which the foodcrops are attached are shown in Figures 13 to 15 for the three nuclides in the decay chain. The one layer soil model represents the reference case (solid line) and comparison is made with the two-layer soil model (broken line). Clearly, reduced radionuclide activity is observed in the top soil in the case of the two-layer soil model. These results are in full agreement with the conclusions drawn from the previous sections.

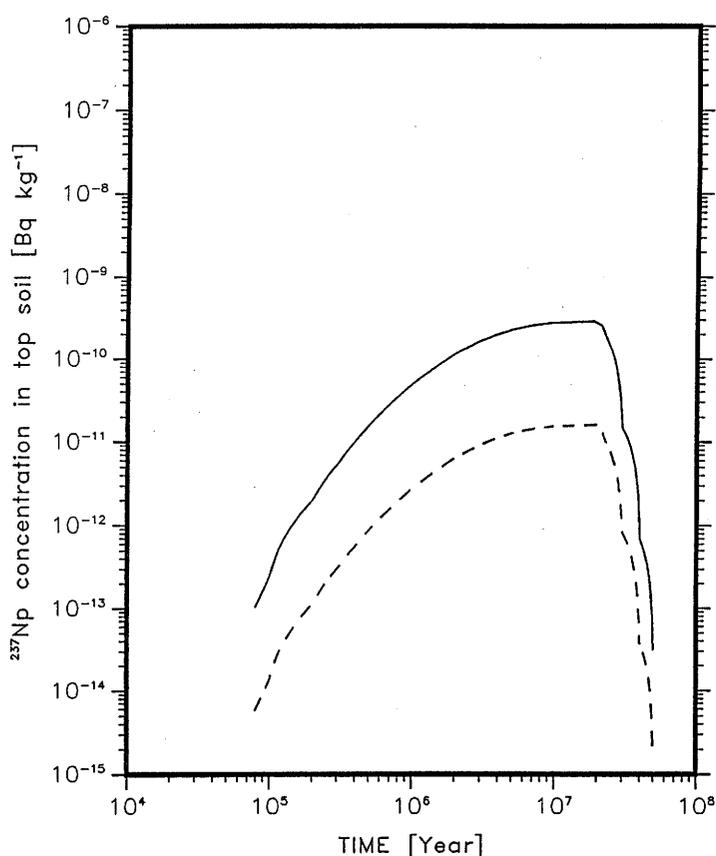


Figure 13: Concentration of  $^{237}\text{Np}$  in root zone soil for a one-layer soil model (solid line) and a two-layer soil model (broken line)

The factor by which the maximum radionuclide concentration (i.e. after about  $10^7$  years) decreases in the top soil for  $^{237}\text{Np}$ ,  $^{233}\text{U}$  and  $^{229}\text{Th}$  compared with the reference case are

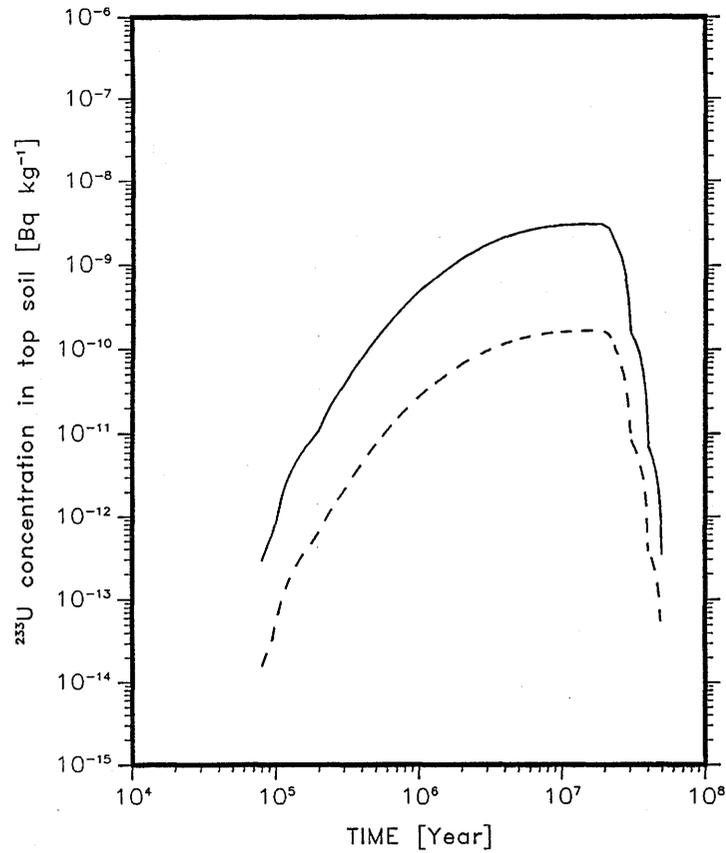


Figure 14: Concentration of  $^{233}\text{U}$  in root zone soil for a one-layer soil model (solid line) and a two-layer soil model (broken line)

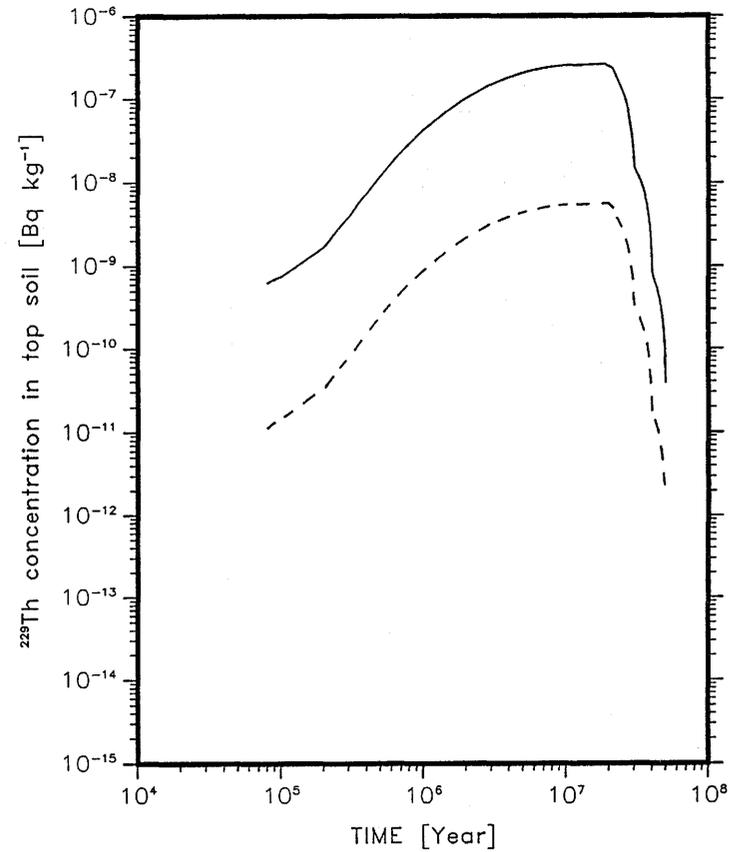


Figure 15: Concentration of  $^{229}\text{Th}$  in root zone soil for a one-layer soil model (solid line) and a two-layer soil model (broken line)

18.0, 18.0 and 46.8, respectively. The lower top soil concentrations are explained by the lower nuclide transfers to this compartment. The difference is most pronounced for thorium because of the very high  $K_d$  value adopted for this element (i.e.  $K_d = 10 \text{ m}^3 \text{ kg}^{-1}$ ).

### 5.2.2 Effect on radiation doses to man

The effect of including a two-layer soil model on the time dependent doses for the individuals (in  $\text{Sv a}^{-1}$ ) for the three nuclides from the neptunium chain are shown in Figure 16. The solid lines represent the reference case (Project Gewähr Study, 1985) while the broken lines show the individual doses as calculated using a two layer soil model. Clearly, there is practically no change in dose for  $^{233}\text{U}$ , a small decrease is observed for  $^{237}\text{Np}$  while a more pronounced decrease occurs for  $^{229}\text{Th}$ . The total individual dose decreases by a factor of 3.2, 1.2 and 1.03 for  $^{229}\text{Th}$ ,  $^{237}\text{Np}$  and  $^{233}\text{U}$  respectively, when a two layer soil model is considered compared with a single layer soil model. Since the root zone soil concentrations are lower in a two layer soil model, and the foodchain pathways are directly attached to this compartment, the final doses are smaller than in the reference case. The reason that the effect is more pronounced for thorium is a reflection of the relatively lower top soil concentrations compared with the other nuclides. These results are summarised further in Table 15.

Table 15: Distribution of nuclide ingestion doses over various exposure paths for the reference case (one-layer soil model) and a two-layer soil model. The values are expressed as % of the total dose for each nuclide of the chain.

Exposure pathway	One-layer soil model			Two-layer soil model		
	$^{237}\text{Np}$	$^{233}\text{U}$	$^{229}\text{Th}$	$^{237}\text{Np}$	$^{233}\text{U}$	$^{229}\text{Th}$
Drinking water	84	96	30	98.9	98.9	95
Milk	-	1	-	-	0.5	-
Meat	-	-	1	0.1	0.1	0.1
Leaf vegetables	1	-	6	0.1	-	0.4
Root vegetables	13	1	35	0.8	0.1	2.4
Cereals	2	2	28	0.1	0.1	1.9
Eggs	-	-	-	-	0.3	0.3
Fish	-	-	-	-	-	-

Table 15 compares the contribution of individual exposure pathways for these nuclides at the time of its dose maximum for the two scenarios. This table shows that the contribution of the drinking water pathway to the total dose for individuals in the reference

case amounts to 96%, 84% and 30% for  $^{233}\text{U}$ ,  $^{237}\text{Np}$  and  $^{229}\text{Th}$ , respectively. Thus, with the exception of thorium, the main pathway resulting in dose to man is drinking contaminated groundwater. In the case of a two layer soil model the contribution of the soil-crop-man pathways to total radiation dose is considerably lower. Therefore, the relative importance of drinking water is further enhanced. For neptunium and uranium this pathway accounts for about 99% of the dose while for thorium an increase from 30% to 95% is observed. On the other hand this table illustrates that the contribution from the food pathway (especially vegetables and cereals) is decreased by factors between 10 and 20. These decreases have a greater effect on the total dose for thorium (less for neptunium and uranium) since these foodchain pathways are of more importance for this element (see Figure 16). It should be realized that here only groundwater to soil and drinking water exposure pathways are compared. The influence of irrigation is discussed in an other study (Grogan and van Dorp, 1986, BIOMOVs, 1989).

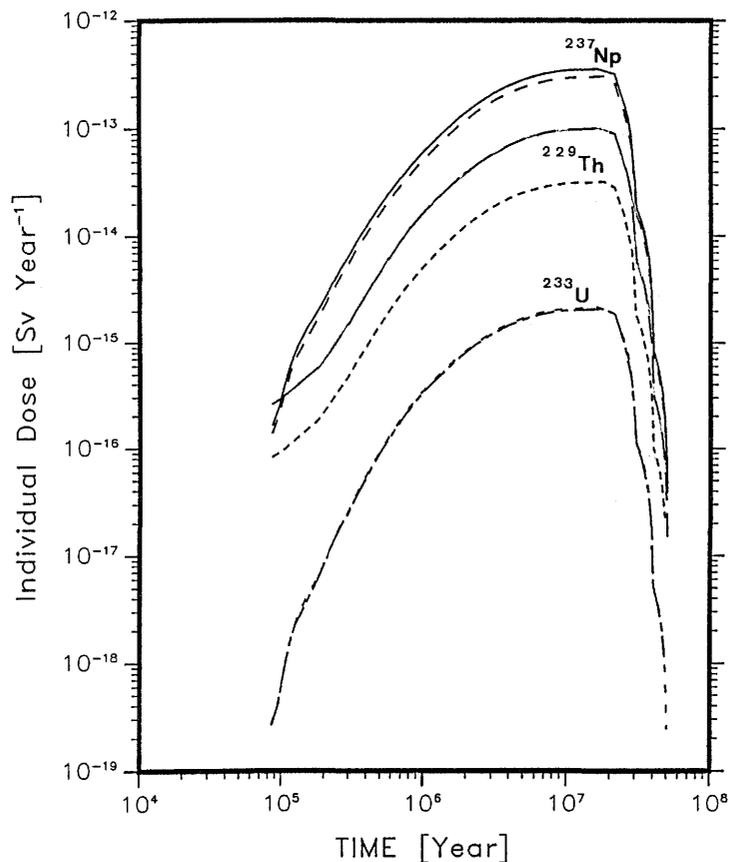


Figure 16: Radiation dose to man for the three nuclides from the neptunium chain for base case (solid line) and for a two-layer soil model (broken line)

## 6 Conclusions

Biosphere modelling for the performance assessment of radioactive waste repositories includes the following steps:

- definition of scenarios
  - e.g. for the nuclide release from the geosphere
  - for the nuclide transfer processes through the biosphere
  - for the exposure pathways to man
- definition of the conceptual model of the biosphere
- formulation of the computer code(s)
- calculations
- interpretation of the results

In this report the release of radioactivity into the biosphere is defined to be via groundwater. The nuclide transfer through the biosphere occurs from a contaminated aquifer to the top soil via a deep soil layer. The only exposure pathway studied is the soil-crop-man pathway. The relative importance of this exposure pathway is related only to a conservative drinking water pathway (which is not varied here) and not with other exposure pathways such as the groundwater-irrigation-crop-man pathway (see NAGRA 1985, BIOMOVIS 1989). The latter would significantly increase the contribution of the soil-crop-man pathway to the total doses but, obviously, this was explicitly excluded from the definition of the scenario in order to assess the importance of upward movement of groundwater to soil.

With respect to this study the following conclusions can be drawn.

- The modelling of nuclide transport from groundwater to rooting zone soil with two soil layers is more realistic than the modelling with one soil layer, because groundwater will rarely flow directly in the top soil in an agriculturally productive area.
- The radionuclide concentration in the top soil layer calculated with two soil layers in the model is lower than calculated with only one soil layer.

- The amount of activity which accumulates in the top soil is determined by (i) the concentration in the groundwater, (ii) the magnitude of the upward flux to the surface soil (iii) the sorption on the soil and (iv) the dilution in the deep soil and the top soil with other water sources such as precipitation surplus and lateral inflowing water.
- The time required to achieve a steady-state concentration in the top soil is determined by the amount of water which flows from the deep soil to the top soil and the extent of radionuclide sorption in both soil layers.

Given a certain input rate of radioactivity into the biosphere at a specified location, uncertainty in calculated soil concentrations is caused by:

- the climatic factors,
- the compartment sizes,
- the water fluxes,
- the sorption of nuclides onto the soil.

Of these parameters, the sorption of nuclides onto the soil shows the largest uncertainty and causes the largest uncertainty in the calculated soil concentrations.

Assuming an inverse relationship between sorption onto the soil and the transfer of activity from soil-to-plant, the uncertainty in the crop concentrations is smaller than that in the soil.

The contribution of the groundwater-soil-crop-man pathway to the total dose to man is reduced when using the two-layer soil concept, compared to the one layer soil concept. The comparison with Project Gewähr 1985 calculations, showed the apparent increase in importance of the drinking water pathway. However, this partly reflects the conservative assumptions made with respect to this pathway. All drinking water is assumed to be from the contaminated groundwater with no processing or filtration of the groundwater. Furthermore no irrigation of crops is assumed, which would significantly increase the contribution to dose for this pathway. This highlights the bias that can result if sections of the conceptual model are improved (made more realistic) in a piecewise manner.

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## 8 References

**AECL 1985**, Second Interim Assessment of the Canadian Concept for Nuclear Fuel-Waste Disposal, Vol.4, Post-Closure Assessment, AECL-8373-4, p.80, Whiteshell Nuclear Research Establishment, Pinawa, Canada, 1985.

**Baes III, C.F.**, Environmental Transport and Monitoring, 1. Prediction of Radionuclide  $K_d$  Values from Soil-Plant Concentration Ratios, Trans. Am. Nucl. Soc., Vol.41, pp.53-54, 1982.

**Bergström, U., Andersson, K., Sundblad, B.**, Biosphere data base revision, SKB Technical Report series, SKB 86-15, Studsvik Energiteknik AB, Nyköping, Sweden, 1986.

**BIOMOVS 1986**, BIOMOVS Progress Report series, Nr. 1, National Institute of Radiation Protection, Stockholm, Sweden, 1986.

**BIOMOVS 1987**, BIOMOVS Progress Report series, Nr. 4, National Institute of Radiation Protection, Stockholm, Sweden, 1987.

**BIOMOVS 1989**, Scenario B2 Irrigation with contaminated groundwater, BIOMOVS Technical Report series, Nr. 6, Ed. H.A. Grogan, National Institute of Radiation Protection, Stockholm, Sweden, 1989.

**Bundi, A.**, Biosphärentransport von Radionukliden – erste Modellierung anhand eines ausgewählten Beispiels, EIR-Bericht Nr. 503, Würenlingen 1983 and NTB 83-22, NAGRA, Baden, Switzerland, 1983

**Coughtrey, P.J., Jackson, D., Thorne, M.C.**, Radionuclide distribution and transport in terrestrial and aquatic ecosystems, Vol. 3, pp. 322-372, A.A. Balkema, Rotterdam, 1983.

**Coughtrey, P.J., Jackson, D., Thorne, M.C.**, Radionuclide distribution and transport in terrestrial and aquatic ecosystems. A compendium of data, Vol.6, pp.69-74, pp.99-103, A.A. Balkema, Rotterdam, 1985.

**Fleming, A.G.**, Essential Micronutrients II: Iodine and Selenium, in Applied Soil Trace Elements, Ed. B.E. Davies, p.203, John Wiley & Sons Ltd., 1980.

**Grogan, H.A.**, Biosphere Modelling for a HLW Repository - Scenario and Parameter Variations, EIR-Bericht Nr. 561, Würenlingen, 1985 and NTB 85-48, NAGRA, Baden, Switzerland, 1985.

**Grogan, H.A.**, Concentration Ratios for BIOPATH: Selection of the Soil-to-Plant Concentration Ratio Database, EIR-Bericht Nr. 575, Würenlingen, 1985 and NTB 85-16, NAGRA, Baden, Switzerland, 1985.

**Grogan, H.A., van Dorp, F.**, BIOMOVs Test Scenario Model Comparison Using BIOPATH, EIR-Bericht Nr. 599, Würenlingen, 1986 and NTB 86-23, NAGRA, Baden, Switzerland, 1986.

**Henrion, P.N., Monsecour, M., Fonteyne, A., Put, M., de Regge, P.**, Migration of Radionuclides in Boom Clay, Radioactive Waste Management and the Nuclear Fuel Cycle, Vol.6 (3-4), pp.313-359, 1985.

Hydrogeologische Karte der Schweiz, Atlas der Schweiz, Blatt 12: Klima und Wetter, 1972.

**Jaeggli, F., Frei, E.**, Die Evapotranspiration Landwirtschaftlicher Kulturen, in Beiträge zur Geologie der Schweiz - Hydrologie, Nr.25, Die Verdunstung in der Schweiz, pp.43-47, Geographischer Verlag Kümmerly + Frey, Bern, 1978.

**Jiskra, J.**, Database for radionuclide transport in the biosphere - Nuclide-specific and geographic data for Northern Switzerland, EIR-Bericht Nr. 547, Würenlingen, 1985 and NTB 85-15, NAGRA, Baden, Switzerland, 1985.

**Kocher, D.C.**, On the long-term behaviour of iodine-129 in the terrestrial environment, in Environmental Migration of Long-lived Radionuclides, Proc. Symposium, Knoxville, 27-31 July 1981, IAEA-SM-257/56, pp.669-679, IAEA, Vienna, 1982.

**Liu, Y., von Gunten, H.R.**, Migration Chemistry and Behaviour of Iodine Relevant to Geological Disposal of Radioactive Wastes - A Literature Review with a Compilation of Sorption Data, PSI-Bericht Nr. 16, Würenlingen, 1988 and NTB 88-29, NAGRA, Baden, Switzerland, 1988.

**Müller, H.**, Personal Communication, 1989.

NAGRA 1985, Project Gewähr Feasibility Study, Nagra Gewähr Report series, NGB 85-01 to NGB 85-09, Nagra, Baden, Switzerland, 1985.

**Ng, Y.C., Colsher, C.S., Thompson, S.E.,** Soil-to-Plant concentration factors for radiological assessments. Final Report, NUREG/CR-2975, UCID-19463, Lawrence Livermore National Laboratory, Livermore, Canada, 1982.

**Nishita, H., Wallace, A., Romney, E.M., Schulz, R.K.,** Effect of soil type on the extractability of Np-237, Pu-239, Am-241 and Cm-244 as a function of pH. Soil Sci. Soc., Vol.132, pp.25-34, 1981.

**Richard, F., Germann, P.,** Berechnung der Evapotranspiration aus der Wasserbilanz des Durchwurzelten Bodens, in Beiträge zur Geologie der Schweiz - Hydrologie, Nr.25, Die Verdunstung in der Schweiz, pp.77-84, Geographischer Verlag Kümmerly + Frey, Bern, 1978.

**Röjder, B., Bergström, U., Edlund, O., Puigdomenech, I.,** BIOPATH: User's Guide, Studsvik Nuclear, Nyköping, Sweden, 1987-1988.

**Scheffer/Schachtschabel,** Lehrbuch der Bodenkunde, 12., neu bearbeitete Auflage, P. Schachtschabel, H.-P. Blume, G. Brümmer, K.-H. Hartge und U. Schwertmann, Ferdinand Enke Verlag Stuttgart, 1989.

**Sheppard, J.C., Campbell, M.J., Kittrick, J.A., Hardt, T.L.,** Retention of neptunium, americium and curium by diffusible soil particles, Environ. Sci. Technol., Vol.44, pp.680-684, 1979.

**Sheppard, M.I., Vandergraaf, T.T., Thibault, D.H., Keith Reid, J.A.,** Technetium and Uranium: Sorption by and plant uptake from peat and sand, Health Physics, Vol.44, No.6 (June), pp.635-643, 1983.

**Sheppard, M.I.,** Radionuclide Partitioning Coefficients in Soils and Plants and Their Correlation, Health Physics, Vol.49, No.1 (July), pp.106-111, 1985.

**Sheppard, S.C.,** Refining Generic Plant/Soil Concentration Ratios, Canadian Nuclear Society, 2nd Inter. Conf. on Radioactive Waste Management, September 7-11 (1986), Conf. Proc., pp.694-696, Winnipeg, Manitoba, Canada, 1986

**Van Loon, L.,** Kinetic aspects of the soil-to-plant transfer of technetium, Dissertationes de Agricultura, PhD Thesis No.150, Katholieke Universiteit Leuven, Belgium, 1986.

**Whitehead, D.C.,** J. Sci. Fd. Agric., Vol.24, pp.547-556, 1973.