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TECHNICAL REPORT 86-23

BIOMOVS Test Scenario Model Comparison Using BIOPATH

Helen A. Grogan[1)]
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SUMMARY

This report presents the results of the irrigation test scenario, presented in the BIOMOVS intercomparison study, calculated by the computer code BIOPATH. This scenario defines a constant release of Tc-99 and Np-237 into groundwater that is used for irrigation.

The system of compartments used to model the biosphere is based upon an area in northern Switzerland and is essentially the same as that used in Projekt Gewaehr to assess the radiological impact of a high level waste repository.

Two separate irrigation methods are considered, namely ditch and overhead irrigation. Their influence on the resultant activities calculated in the groundwater, soil and different foodproducts, as a function of time, is evaluated.

The sensitivity of the model to parameter variations is analysed which allows a deeper understanding of the model chain. These results are assessed subjectively in a first effort to realistically quantify the uncertainty associated with each calculated activity.

Zusammenfassung

Wir berichten über die Resultate eines mit dem Computercode BIOPATH berechneten Bewässerungsszenarios, welches im Rahmen der internationalen Vergleichsstudie BIOMOVS vorgestellt wurde. Dieses Szenario ist dadurch charakterisiert, dass eine konstant gehaltene Menge von Tc-99 und Np-237 kontinuerlich in das Grundwasser, welches für Bewässerungszwecke verwendet wird, gelangt.

Das System von Kompartimenten, welches für die Modellierung der Biosphäre verwendet wird, basiert auf einem Gebiet der Nordschweiz; es ist im wesentlichen dasselbe wie im Projekt Gewähr, in welchem der radiologische Einfluss eines Endlagers für hochaktive Abfälle untersucht wurde.

Es wurden zwei verschiedene Bewässerungsarten angesehen; einerseits Kanalbewässerung und andererseits Sprinklerbewässerung. Ihr Einfluss auf die resultierenden Aktivitäten im Grundwasser, Boden und verschiedenen Nahrungsprodukten wurde in Funktion der Zeit studiert.

Die Empfindlichkeit des Modells auf Parametervariationen wurde analysiert, um ein besseres Verständnis der Modellkette zu erlangen: Die Resultate dienen dazu, die Unsicherheiten, die den berechneten Aktivitäten anhaften, in einer ersten Anstrengung realistisch abzuschätzen.

RESUME

Ce rapport présente les résultats calculés au moyen du code BIOPATH, du scénario d'irrigation proposé dans le cadre de l'étude internationale de comparaison BIOMOVS. Ce scénario est caractérisé par un relâchement constant de Tc-99 et Np-237 dans les eaux souterraines utilisées pour l'irrigation.

Le système de compartiments utilisé pour la modélisation de la biosphère est basé sur une région du nord de la Suisse et est essentiellement le même que celui qui est utilisé lors du projet "Gewaehr" pour l'étude de l'impact radiologique d'un dépôt final de déchets hautement radioactifs.

Deux méthodes d'irrigation distinctes ont été considérées, notamment au moyen de canaux et au moyen d'installations d'aspersion. Leur influence sur les activités résultantes, calculées en fonction du temps dans les eaux souterraines, le sol et différents aliments, a été évaluée.

La sensibilité du modèle à des variations des paramètres a été anaylsée, ce qui permet une compréhension plus profonde de la chaîne du modèle. Dans une première étape, ces résultats servent à évaluer de façon réaliste les marges d'erreur qui affectent chaque activité calculée.

CONTENTS PAGE

1.INTRODUCTION AND BENCHMARK DEFINITION

BIOMOVS is an international study of models designed to predict the environmental transfer and bioaccumulation of radionuclides and other trace substances. This study is managed by the Swedish National Institution of Radiation Protection (NIRP) with Kemakta Consultants Co. acting as the secretariat.

The primary objectives of the study can be summarised in three points. Firstly, to test the accuracy of the predictions of environmental assessment models for selected contaminants and exposure scenarios. Secondly, to explain the differences in the model predictions. These may be due to structural differences in the models, invalid assumptions and/or differences in selected input data. Finally, it is intended that priorities for future research to improve the accuracy of model predictions can be recommended.

It is anticipated that few problems will arise from the calculational and numerical part of the modelling, since most models/codes are mathematically simple and code verification is easy. However, it is the conceptual basis of the various approaches and the often unstated implicit assumptions that are likely to be the major source of varying model output.

In Switzerland, the computer code BIOPATH /1/ is used to assess the radiological consequences of radionuclide release into the biosphere from an underground repository /2/. This model has therefore been entered into the BIOMOVS study /3/.

Two separate approaches are involved in the BIOMOVS comparison. In approach A, the models are tested against independent data sets. The trace substance concentrations in various parts of the biosphere are calculated as a function of a given concentration in air, surface water or groundwater over a specific period of time. In approach A these quantities will be compared with measured data. However, collation of such data sets takes time and at present none are available for analysis.

For the alternative approach (B), specific test scenarios are defined and the model predictions and associated uncertainties are compared. This approach requires much less preparation since no independent data sets are required and the scenarios can be chosen on the basis of assessment priorities. This approach has therefore been embarked upon first.

To date three different test scenarios for approach B have been defined in BIOMOVS. In the first, the radioactivity is released into the atmosphere, in the second the radioactivity is released into groundwater and in the third it is released into a lake from a river. The scenarios are defined rather loosely so that differences in modelling approaches and structure become visible as well as input data selection for a non site-specific scenario.

In this report the results obtained using BIOPATH to calculate the results of test scenario II are presented. This scenario was chosen for analysis because it is very similar to the situation modelled in Projekt Gewaehr for assessing the radiological impact of a high level waste repository (type C). A description of test scenario II, as specified in BIOMOVS is given below.

<u>Test Scenario II</u> - irrigation with contaminated groundwater.

Given an activity concentration of the radionuclides Np-237 and Tc-99 (1 Bq l^{-1}) in groundwater over a period of 10,000 years

1) Calculate the concentration at times 1, 10, 100, 1000 and 10000 years in the root zone of surface soil for (a) undisturbed pastures and (b) ploughed agricultural land. Assume that this groundwater is used as a source of spray irrigation and that the average rate of irrigation is 300 mm per year during a 6 months growing season.

2) Calculate the concentrations at times 1, 10, 100, 1000 and 10000 years in the edible portions of pasture forage (Bq kg^{-1}, dry wt), milk (Bq l^{-1}), and meat (Bq kg^{-1}, fresh wt), and in leafy vegetables (Bq kg , fresh wt), grains (Bq kg^{-1}, fresh wt), and root crops (Bq kg^{-1}, fresh wt) prior to human consumption.

3) Calculate the concentration in the above-ground atmosphere (in Bq or mg m^{-3}) resulting from resuspension from the soil.

The series of physical compartments making up the biosphere model (Fig. 1) are based upon the Laufenburg region in northern Switzerland. The Laufenburg region is situated on a broad gravel terrace along the south bank of the Rhine river. This system is essentially the same as that used to assess the radiological impact of an underground repository /2/,/4/.

In this study the two daughter nuclides of Np-237 namely, U-233 and Th-229, were also considered. In the base case the Np-237 activity was set at 1 Bq l^{-1} in the groundwater and the two daughters grew in with time. A second variation was studied in which the daughters also had an activity concentration of 1 Bq l^{-1} in the groundwater .

The half life of each radionuclide modelled is listed below.

Table 1: Radionuclide Half - Lives.

Radionuclide	Half-life (a)
Tc-99	2.13×10^{5}
Np-237	2.14×10^{6}
U-233	1.59×10^{5}
Th-229	7.34×10^{3}

Two separate methods for irrigation were considered :

a) ditch irrigation : Contaminated irrigation water is applied to the soil in ditches, so that crop contamination occurs via root uptake.

b) overhead irrigation :Contaminated irrigation water is applied by sprinklers so that crops are both directly (via foliage) and indirectly (via root uptake) contaminated.

No calculations were made for the first time period (1 year) because the model is designed for long term assessment. In addition the model does not consider timesteps less than one year so that although the test scenario specifies irrigation over a 6 month period this is automatically integrated over one year. In the model the soil is simply treated as a homogeneous 25 cm deep layer. No distinction is made between ploughed and unploughed soil because, for long term assessment (>100years) these differences are negligible.

2. MODEL DESCRIPTION

Model BIOPATH

Purpose of Model

The model is used as an assessment tool for determining the radiological consequences of potential releases of radionuclides from an underground repository.

It is designed to demonstrate compliance with regulatory standards promulgated to protect the individual (population). In Switzerland these are currently framed in terms of an annual individual committed dose equivalent (10 mrem a^{-1}).

Intended Accuracy of Model Predictions

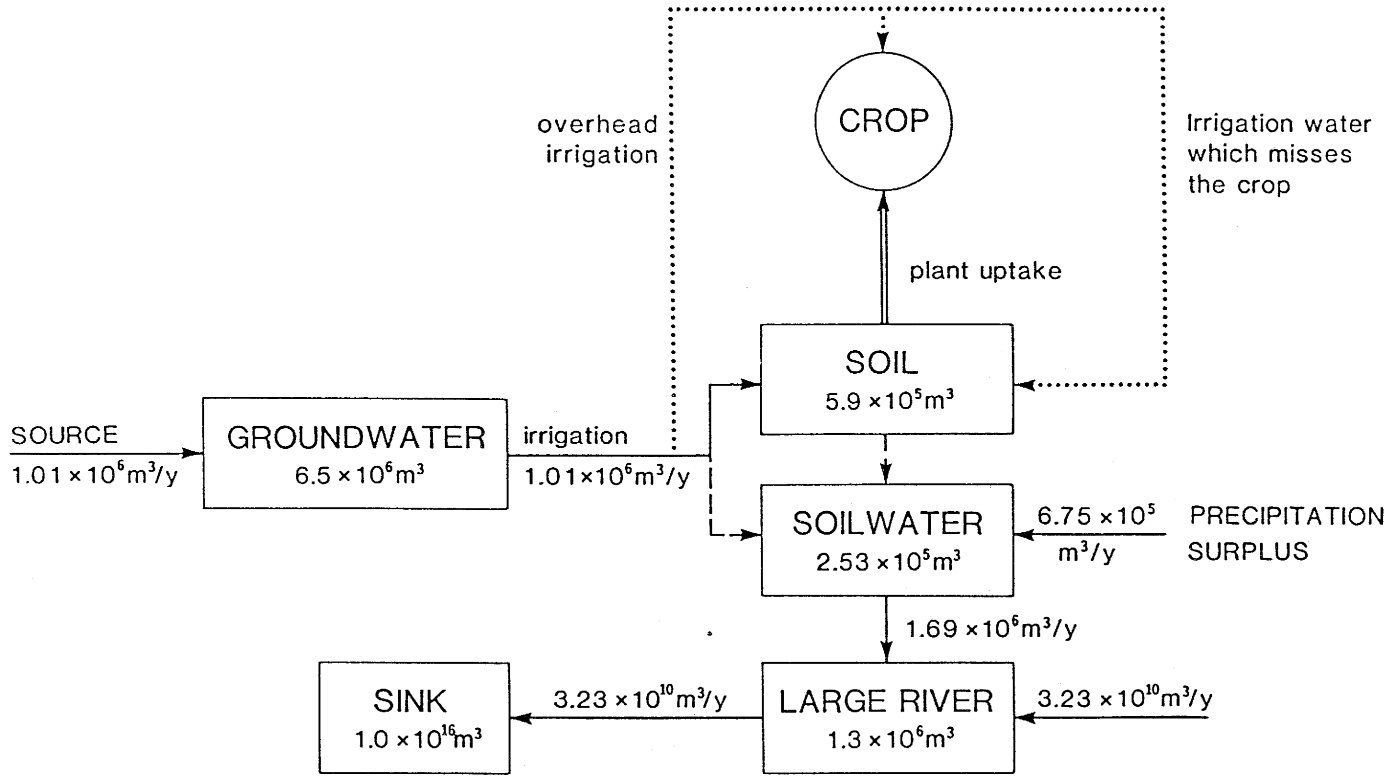
The model predictions should represent a conservative bias in order to avoid a substantial underestimation of contaminant concentrations or radiological consequences. This is due to the basic uncertainties in the parameters and their extrapolation over long timescales.

Model Structure

The biosphere model is divided into a series of physical compartments through which the radionuclide flow is calculated as a function of time.

This dynamic model leads to a linear system that is described by coupled first order ordinary differential equations. The foodchain pathways are attached to the appropriate physical compartments /1/. This foodchain portion only requires simple steady-state transfer factors because equilibrium is assumed among concentrations in the different foodchain compartments. In the model the crops are divided into four seperate categories, pasture, cereal (grain), leaf vegetables and root vegetables.

Figure 1 shows the physical compartments used to model the irrigation scenario along with the fluxes between the compartments. Both irrigation pathways are indicated in the diagram : i) ditch irrigation, ii) overhead irrigation.



key

$6.5 \times 10^6 m^3$	compartment volume
$5.5 \times 10^6 m^3/y$ ⟶	waterflux
– – – – – – ➤	radionuclide distribution

Note:

Irrigation can either be overhead or directly onto the soil (with subsequent root uptake).

Fig. 1: Compartments and Water Fluxes for the Irrigation Scenario

Model Input

Radionuclide concentration as a function of time in the source term.

Volume and density of each compartment in the model chain.

Transfer coefficients specifying the transfer rate of each radionuclide between the compartments.

Radionuclide specific parameters eg, decay constants, concentration ratios, distribution factors, dose conversion factors etc.

Consumption values (animal / human) and living habits.

Population size.

Model Results (Output)

The results of the model are: -

- nuclide concentration in each of the physical compartments as a function of time.
- nuclide concentrations in the foodchain products.
- individual/population doses as a function of time.

Calculation of Radionuclide Activity in Foodcrops and Animal Produce

In this section the method by which the radionuclide activity in a crop or animal product is calculated, is presented. This differs slightly depending whether ditch or overhead irrigation is assumed so one example of each is given. For full details see reference /14/. In the next chapter the individual parameter values are specified along with a discussion of their variability.

Radionuclide activity in green vegetables is given by the following expressions :-

i) ditch irrigation

$$C_s \cdot CR_{gv}$$

where C_s = radionuclide activity in soil (Bq kg^{-1})
CR_{gv} = soil - green vegetable concentration ratio

ii) overhead irrigation

$$C_s \cdot CR_{gv} + \frac{N \cdot T_v \cdot W \cdot C_w}{Y}$$

N = retention factor
T_v = retention half-life (d)
W = irrigation rate (l m^{-2} d^{-1})
C_w = activity in irrigation water (Bq l^{-1})
Y = crop density (kg m^{-2})

Radionuclide activity in milk is given by the following expressions :-

i) ditch irrigation

$$C_s \cdot CR_{past} \cdot Con_{past} + xC_s + Con_w \cdot C_w$$

Con_{past} daily pasture consumption (kg (fresh wt.) d^{-1})
Con_w daily water consumption (l d^{-1})
x daily soil ingestion (kg d^{-1})

ii) overhead irrigation

$$C_s \cdot CR_{past} + \frac{N \cdot T_v \cdot W \cdot C_w}{Y} Con_{past} + xC_s + Con_w \cdot C_w$$

In calculating the activity in milk it is assumed that cows also ingest some soil whilst grazing (see page 10).

3. DATA SOURCES AND UNCERTAINTY ESTIMATES

Compartment Volumes and Fluxes

The compartment volumes and fluxes are based on the Laufenburg region in northern Switzerland, the physical area of which is calculated from topographical maps.

Groundwater: The groundwater volume is calculated from hydrogeological maps of the region which indicate the depth and extent of the aquifer. The flow porosity of the rock is assumed to be 20%. For the purposes of this modelling exercise the groundwater flux is simply calculated as the surface area of the region multiplied by the annual rate of irrigation (300 mm m^{-2}). Under normal circumstances however, the flux is calculated from the slope of the groundwater surface, the hydraulic permeability and the average cross sectional area of the region.

Soil/Soil Water: The soil is assumed to be 25 cm deep, covering the entire area. The soil is homogeneous throughout and is subdivided into soil solid and soilwater which make up 70% and 30% by volume, respectively. The Kd concept is used to model the partitioning of the radionuclides between these two compartments. The Kd values ($m^3\ kg^{-1}$) range from 10^{-3} to 10 and are quoted to the nearest order of magnitude for each element (cf Table 15 following). The default values are based upon a literature review by J. Jiskra /5/.

Foodchain Data: The concentration ratios (CR) selected /6/ were based on a comprehensive literature review in which emphasis was placed upon the more recent experimental work carried out under well defined field conditions. In most instances the values reported in the literature are on a dry weight basis so the following crop water contents were assumed for conversion into fresh plant weight CR's.

Table 2: Crop Moisture Contents.

Crop	% Water Content
Pasture	75
Cereals	11
Leaf vegetables	90
Root vegetables	85

Default CR's Used in the Model and Uncertainty Estimates

The default CR's along with the minimum and maximum value estimates used in the model are expressed as the radionuclide concentration per unit mass fresh plant divided by the radionuclide concentration per unit mass soil (dried weight).

Technetium

Table 3: Concentration Ratios For Technetium.

Tc-99	Default CR	Maximum Value	Minimum Value
Pasture	2.5	2.5×10^{1}	2.5×10^{-2}
Cereals	4.5	4.5×10^{1}	5.0×10^{-3}
Leaf Veg.	1.0	1.0×10^{1}	1.0×10^{-2}
Root Veg.	1.5	1.5×10^{1}	1.5×10^{-2}

These default values are taken from short term field and laboratory experiments (single season) and are probably conservative for the long term situation. However very few long term field experiments have been carried out with Tc-99. Eriksson /7/ demonstrated a decrease in CR by a factor of 2000 for Tc-99 over five years. For this reason the minimum values are two or more orders of magnitude less than the default values, whereas the maximum values are not expected to be more than 10 times geater.

It should be noted that, in the model, the CR input values are not correlated to the Kd input values.

Neptunium

Table 4: Concentration Ratios For Neptunium.

Np-237	Default CR	Maximum Value	Minimum Value
Pasture	9.4×10^{-3}	1.4×10^{-1}	6.3×10^{-4}
Cereals	1.7×10^{-2}	2.5×10^{-1}	4.5×10^{-5}
Leaf Veg.	2.7×10^{-2}	1.3×10^{-1}	4.5×10^{-4}
Root Veg.	6.0×10^{-2}	3.0×10^{-1}	3.0×10^{-4}

The Np-237 CR values were mainly derived from the work of Eriksson /8/, Schreckhise and Cline /9/, and Pimpl and Schmidt /10/. The default CR values are the arithmetic means of the experimentally measured CR's. In the case of root vegetables no experimental values could be found so a CR equivalent to 0.4 (dry plant) as recommended by Schreckhise and Cline, as a result of their findings with other crop-types, was adopted. The minimum and maximum values are taken from the extremes of the reported experimental measurements. It should be added that Pimpl has recorded a CR of 5.7×10^{-7} (dry plant) for pasture although this was not included as the minimum expected value since it is significantly less than any other reported value. However, this may suggest that the minimum values could all be at least one order of magnitude less than those stated, indicating less importance of the root-uptake pathway.

Uranium and Thorium

Table 5: Concentration Ratios For Uranium And Thorium.

U-233 Th-229	Default CR	Maximum Value	Minimum Value
Pasture	9.5×10^{-4}	1.4×10^{-2}	6.3×10^{-5}
Cereal U-233	1.3×10^{-3}	2.0×10^{-2}	8.7×10^{-5}
Th-229	7.1×10^{-4}	1.1×10^{-2}	4.7×10^{-5}
Leaf veg.	3.8×10^{-4}	5.7×10^{-3}	2.5×10^{-5}
Root veg.	5.7×10^{-4}	8.6×10^{-3}	3.8×10^{-5}

There are few experimental determinations for U and Th uptake by crops in the literature. Evans and Eriksson /11/ have determined U and Th CR's for crops grown at 37 different sites in Sweden and the default values are based on the arithmetic means of these results. The mean CR's for U and Th are the same in all the crop types studied, except for cereal grain. In this case both elements showed a lower uptake into cereal with the U being slightly preferentially taken up. Neither element appears to demonstrate a large fluctuation in uptake therefore a factor 15 was arbitrarily chosen as an uncertainty estimate for both the minimum and maximum values.

Distribution Factors

Distribution factors (DF) are used to define the animal concentration ratios and are best exemplified by the DF for milk.

$$DF = \frac{\text{Bq } l^{-1} \text{ milk}}{\text{Bq kg}^{-1} \text{feed} \;.\; \text{kg intake feed d}^{-1}}$$

The activity of the radionuclide in the milk is assumed to be in equilibrium with the concentration in the feed. The distribution factors for milk and meat are documented in /5/ and were mainly derived from Ng's literature review /12/.

Table 6: Milk And Meat Distribution Factors.

Element	Milk ($d\ l^{-1}$)	Meat ($d\ kg^{-1}$)
Tc	2.5×10^{-2}	1.0×10^{-3}
Np	5.0×10^{-6}	2.0×10^{-4}
U	3.7×10^{-4}	3.4×10^{-4}
Th	5.0×10^{-6}	2.0×10^{-4}

Animal Consumption

The animal consumption data were obtained from Swiss agricultural statistics.

Table 7: Cattle Consumption Data.

	Water	Fodder (wet weight)
Average daily consumption by cattle	30 l	100 kg

Soil Ingestion By Cattle

When calculating the radionuclide activity in milk and meat it is assumed that the livestock also ingest some soil whilst grazing the pasture. A value of 4% soil ingestion by weight of the daily dry matter intake is taken /13/. This is equivalent to 1kg dry soil per animal per day for an intake of 100kg fodder (fresh weight) as assumed in BIOPATH.

Soil Resuspension

This is the process by which the contaminated soil particles become inserted into the atmosphere as a result of wind or mechanical stresses. Most data on resuspension have been reported in the form of a resuspension factor (K) defined as :-

$$K = \frac{\text{resuspension air concentration}}{\text{surface concentration}} \quad m^{-1}$$

Experimentally determined values of K range over more than seven orders of magnitude and for a more complete description see /14/,/15/,/16/.

Resuspension is not actually included in the BIOPATH code at present. Consequently at this stage resuspension has been considered rather briefly, whereby only a maximum air concentration has been calculated. This was assumed as 50 µg of contaminated soil per m^3 air. This is equivalent to a resuspension rate (K) of 2.2×10^{-8} m^{-1} which lies at the upper end of the range of experimentally determined values for 'aged' deposits /14/.

Overhead Irrigation Parameters

In overhead irrigation contaminated irrigation water falls directly onto the plant foliage and can thus enter the crop. The activity content of the plant resulting from this route is calculated according to the following equation and a more complete account of this is given in /17/.

$$C_p = \frac{N \cdot T_v \cdot W \cdot C_w}{Y}$$

N = retention factor
T_v = retention half-life (d)
W = irrigation rate ($l\ m^{-2}\ d^{-1}$)
C_w = activity in irrigation water (Bq l^{-1})
Y = crop density (kg m-2)
C_p = activity in plant (Bq kg^{-1})

Table 8: Specification of the Overhead Irrigation Parameter Values.

Crop	N	T (d)	Y (kg m^{-2})	Source
pasture	0.3	14	0.85 (per cut)	/18/,/19/,/22/
cereal	0.05	30	0.50	/18/,/7/,/23/ /20/,/21/
leaf veg.	0.3	14	2.20	/20/,/24/,/25/,/26/

These parameter values have been taken from experiments designed to study foliar uptake subsequent to a single application of radionuclides. Very little work has been carried out explicitly on the extent of foliar uptake of radionuclides as a result of irrigation. For this reason no uncertainty estimates are offered for these parameters.

In fact, it is considered that a series of greenhouse experiments that examine radionuclide retention and uptake by crops during irrigation, would be of great value for the modelling. It is only subsequent to such investigations that any real estimates of parameter uncertainties can be made.

4. TEST SCENARIO II RESULTS

The base case results for the irrigation scenario are presented in the following tables. In the base case the default values for all input parameters are taken. In the base case the Tc-99 and Np-237 enter the system at a constant activity of 1 Bq l^{-1} and the irrigation is at a rate of 300 mm a^{-1}. Tables 10 and 11 give the radionuclide activity as a function of time, in the soil and groundwater, respectively. In table 12 the temporal variation in radionuclide activity in the different food crops and products as a result of ditch irrigation, are presented. For comparison table 13 presents the corresponding activities when overhead irrigation is assumed.

The factor by which the minimum and maximum values are expected to deviate from the calculated values are also indicated. A discussion of how these values were selected is given in the next section, under "Parameter Variations". The table below presents the concentration of the radionuclides in the air resulting from resuspension. This is a maximum value for the base case situation and is obtained by assuming a resuspension rate of 50 µg of soil per m^3 of air.

Table 9: Radionuclide Concentration in the Air as a Result of Resuspension (Bq m^{-3}).

Nuclide	Time (a)			
	10	100	1000	10000
Tc-99	$2.1x10^{-8}$	$3.0x10^{-8}$	$3.0x10^{-8}$	$3.0x10^{-8}$
Np-237	$1.1x10^{-7}$	$3.0x10^{-7}$	$3.0x10^{-7}$	$3.0x10^{-7}$
U-233	$3.3x10^{-12}$	$1.2x10^{-10}$	$1.9x10^{-10}$	$1.9x10^{-10}$
Th-229	$1.1x10^{-15}$	$6.5x10^{-13}$	$1.5x10^{-11}$	$1.0x10^{-10}$

Table 10 : Radionuclide concentration in the root zone of the surface soil (Bq kg^{-1} dry weight)

	10 years			100 years			1000 years			10000 years		
Radionnuclide	conc.	min	max	conc.	min	max	conc.	min	max	conc.	min	max
Tc-99	0.4	---	7.5	0.6	---	105	0.6	---	450	0.6	---	500
Np-237	2.1	---	1.5	6.0	---	11	6.0	---	71	6.0	---	100
U-233	6.6×10^{-5}	---	1.2	2.4×10^{-3}	---	6.5	3.8×10^{-3}	---	328	3.8×10^{-3}	---	556
Th-229	2.2×10^{-8}	---	1.1	1.3×10^{-5}	---	3.3	3.1×10^{-4}	---	285	2.1×10^{-3}	---	487

Table 11 : Radionuclide concentration in the groundwater (Bq l^{-1})

	10 years			100 years			1000 years			10000 years		
Radionuclide	conc.	min	max	conc.	min	max	conc.	min	max	conc.	min	max
Tc-99	0.7	---	---	1.0	---	---	1.0	---	---	1.0	---	---
Np-237	0.8	---	---	1.0	---	---	1.0	---	---	1.0	---	---
U-233	1.1×10^{-5}	---	---	2.8×10^{-5}	---	---	2.8×10^{-5}	---	---	2.8×10^{-5}	---	---
Th-229	2.8×10^{-9}	---	---	1.7×10^{-8}	---	---	1.7×10^{-8}	---	---	1.7×10^{-8}	---	---

Note :

Min./max. values are expressed as the factor by which they are smaller/greater than the calculated concentration

- No estimates made

Table 12: Radionuclide Concentrations in the Foodchain Products as a Result of Ditch Irrigation (including uncertainty estimates).

		10 years			100 years			1000 years			10000 years		
Product	Radionuclide	conc.	min	max	conc.	min	max	conc.	min	max	conc.	min	max
Pasture forage ($Bq\ kg^{-1}$)	Tc-99	1.3	100	10	1.8	100	90	1.8	---	400	1.8	---	400
	Np-237	$2.0x10^{-2}$	15	15	$5.8x10^{-2}$	15	15	$5.8x10^{-2}$	---	100	$5.8x10^{-2}$	---	100
	U-233	$6.2x10^{-8}$	15	15	$2.3x10^{-6}$	15	15	$3.6x10^{-6}$	---	560	$3.6x10^{-6}$	---	560
	Th-229	$2.1x10^{-11}$	15	15	$1.2x10^{-8}$	15	15	$2.9x10^{-7}$	---	285	$2.0x10^{-5}$	---	495
Milk ($Bq\ kg^{-1}$)	Tc-99	3.7	100	10	5.3	100	75	5.3	---	350	5.3	---	350
	Np-237	$1.3x10^{-4}$	15	15	$2.1x10^{-4}$	15	15	$2.1x10^{-4}$	---	20	$2.1x10^{-4}$	---	20
	U-233	$1.5x10^{-7}$	15	15	$1.8x10^{-8}$	15	15	$1.8x10^{-6}$	---	170	$1.8x10^{-6}$	---	170
	Th-229	$5.5x10^{-13}$	15	15	$7.3x10^{-11}$	15	15	$1.7x10^{-9}$	---	280	$1.\ x10^{-8}$	---	280
Meat ($Bq\ kg^{-1}$)	Tc-99	$1.5x10^{-1}$	100	10	$2.1x10^{-1}$	100	75	$2.1x10^{-1}$	---	350	$2.1x10^{-1}$	---	350
	Np-237	$5.3x10^{-3}$	15	15	$8.3x10^{-3}$	15	15	$8.3x10^{-3}$	---	20	$8.3x10^{-3}$	---	20
	U-233	$1.4x10^{-7}$	15	15	$1.2x10^{-6}$	15	15	$1.7x10^{-6}$	---	170	$1.7x10^{-6}$	---	170
	Th-229	$2.2x10^{-11}$	15	15	$2.9x10^{-9}$	15	15	$6.8x10^{-8}$	---	280	$4.5x10^{-7}$	---	280
Leaf Vegetables ($Bq\ kg^{-1}$)	Tc-99	$5.0x10^{-1}$	100	10	$7.2x10^{-1}$	100	90	$7.2x10^{-1}$	---	400	$7.2x10^{-1}$	---	400
	Np-237	$5.8x10^{-2}$	90	5	$1.7x10^{-1}$	90	5	$1.7x10^{-1}$	---	100	$1.7x10^{-1}$	---	100
	U-233	$2.5x10^{-8}$	15	15	$9.3x10^{-7}$	15	15	$1.4x10^{-6}$	---	560	$1.4x10^{-6}$	---	560
	Th-229	$8.4x10^{-12}$	15	15	$4.9x10^{-9}$	15	15	$1.2x10^{-7}$	---	285	$7.9x10^{-7}$	---	285
Grain ($Bq\ kg^{-1}$)	Tc-99	2.3	100	10	3.2	100	90	3.2	---	400	3.2	---	400
	Np-237	$3.6x10^{-2}$	900	15	$1.0x10^{-1}$	900	15	$1.0x10^{-1}$	---	100	$1.0x10^{-1}$	---	100
	U-233	$8.5x10^{-8}$	15	15	$3.2x10^{-6}$	15	15	$4.9x10^{-6}$	---	560	$4.9x10^{-6}$	---	560
	Th-229	$1.6x10^{-11}$	15	15	$9.2x10^{-9}$	15	15	$2.2x10^{-7}$	---	285	$1.5x10^{-6}$	---	285
Root Crops ($Bq\ kg^{-1}$)	Tc-99	$7.5x10^{-1}$	100	10	1.1	100	90	1.1	---	400	1.1	---	400
	Np-237	$1.3x10^{-1}$	130	5	$3.7x10^{-1}$	130	5	$3.7x10^{-1}$	---	100	$3.7x10^{-1}$	---	100
	U-233	$3.7x10^{-8}$	15	15	$1.4x10^{-6}$	15	15	$2.1x10^{-6}$	---	560	$2.1x10^{-6}$	---	560
	Th-229	$1.3x10^{-11}$	15	15	$7.3x10^{-9}$	15	15	$1.8x10^{-7}$	---	285	$1.2x10^{-6}$	---	285

Note: min./max. values are expressed as the factor by which they are smaller/greater than the calculated concentration

Table 13: Radionuclide Concentrations in Foodchain Products as a Result of Overhead Irrigation (including uncertainty estimates).

		10 years			100 years			1000 years			10000 years		
Product	Radionuclide	conc	min	max	conc	min	max	conc	min	max	conc	min	max
Pasture Forage ($Bq\ kg^{-1}$)	Tc-99	4.3	100	10	5.9	100	90	5.9	---	400	5.9	---	400
	Np-237	3.0	15	15	4.1	15	15	4.1	---	100	4.1	---	100
	U-233	4.6×10^{-5}	15	15	1.2×10^{-4}	15	15	1.2×10^{-4}	---	560	1.2×10^{-4}	---	560
	Th-229	1.1×10^{-8}	15	15	8.1×10^{-8}	15	15	3.6×10^{-7}	---	285	2.0×10^{-6}	---	495
Milk ($Bq\ kg^{-1}$)	Tc-99	11.3	100	10	15.4	100	75	15.4	---	350	15.4	---	350
	Np-237	1.7×10^{-3}	15	15	2.2×10^{-3}	15	15	2.2×10^{-3}	---	20	2.2×10^{-3}	---	20
	U-233	1.9×10^{-6}	15	15	5.5×10^{-6}	15	15	6.1×10^{-6}	---	170	6.1×10^{-6}	---	170
	Th-229	6.3×10^{-12}	15	15	1.1×10^{-10}	15	15	1.7×10^{-9}	---	280	1.1×10^{-8}	---	280
Meat ($Bq\ kg^{-1}$)	Tc-99	4.5×10^{-1}	100	10	6.2×10^{-1}	100	75	6.2×10^{-1}	---	350	6.2×10^{-1}	---	350
	Np-237	6.7×10^{-2}	15	15	9.0×10^{-2}	15	15	9.0×10^{-2}	---	20	9.0×10^{-2}	---	20
	U-233	1.7×10^{-6}	15	15	5.1×10^{-6}	15	15	5.6×10^{-6}	---	170	5.6×10^{-6}	---	170
	Th-229	2.5×10^{-10}	15	15	4.3×10^{-9}	15	15	7.0×10^{-8}	---	280	4.5×10^{-7}	---	280
Leaf Vegetables ($Bq\ kg^{-1}$)	Tc-99	1.7	100	10	2.3	100	90	2.3	---	450	2.3	---	400
	Np-237	1.2	90	5	1.7	90	5	1.7	---	100	1.7	---	100
	U-233	1.8×10^{-5}	15	15	4.5×10^{-5}	15	15	4.5×10^{-5}	---	560	4.5×10^{-5}	---	560
	Th-229	4.4×10^{-9}	15	15	3.2×10^{-8}	15	15	1.5×10^{-7}	---	285	8.1×10^{-7}	---	285
Grain ($Bq\ kg^{-1}$)	Tc-99	4.1	100	10	5.7	150	90	5.7	---	400	5.7	---	400
	Np-237	1.9	900	15	2.6	900	15	2.6	---	100	2.6	---	100
	U-233	2.8×10^{-5}	15	15	7.2×10^{-5}	15	15	7.4×10^{-5}	---	560	7.4×10^{-5}	---	560
	Th-229	7.0×10^{-9}	15	15	5.1×10^{-8}	15	15	2.6×10^{-7}	---	285	1.5×10^{-6}	---	285
Root Crops ($Bq\ kg^{-1}$)	Tc-99	7.5×10^{-1}	100	10	1.1	150	90	1.1	---	400	1.1	---	400
	Np-237	1.3×10^{-1}	130	5	3.7×10^{-1}	130	5	3.7×10^{-1}	---	100	3.7×10^{-1}	---	100
	U-233	3.7×10^{-8}	15	15	1.4×10^{-6}	15	15	2.1×10^{-6}	---	560	2.1×10^{-6}	---	560
	Th-229	1.3×10^{-11}	15	15	7.3×10^{-9}	15	15	1.8×10^{-7}	---	285	1.2×10^{-6}	---	285

Note: min./max. values are expressed as the factor by which they are smaller/greater than the calculated concentration

5. PARAMETER VARIATIONS AND RESULTS

The sensitivity of the model to parameter variations was analysed to allow a better understanding of the model chain. These results were then used to realistically estimate the uncertainty associated with calculated results. The various parameter variations are described below.

Effect of Altering the Rainfall Regime

In this case the irrigation rate was kept constant at 300 mm a^{-1}, but the extent of precipitation surplus was varied around the base case.

a) Base Case
300 mm a^{-1} irrigation, 200 mm a^{-1} excess precipitation.

b) Variation 1 (low rainfall)
300 mm a^{-1} irrigation, no excess precipitation.

c) Variation 2 (high rainfall)
300 mm a^{-1} irrigation, 400 mm a^{-1} excess precipitation.

The results are shown in the table below as the factor by which the calculated soil activity deviates from that in the base case.

Table 14: Results of Varying the Rainfall Regime.

Nuclide	Case	Time (a) 10	100	1000	10000
Tc-99	V1	1.59	1.70	1.70	1.70
	V2	0.73	0.71	0.71	0.71
Np-237	V1	1.15	1.66	1.67	1.67
	V2	0.88	0.72	0.72	0.72
U-233	V1	1.05	1.59	2.28	2.28
	V2	0.94	0.72	0.60	0.60
Th-229	V1	1.04	1.43	2.15	2.67
	V2	0.96	0.78	0.60	0.45

V1 = variation 1 (low rainfall)
V2 = variation 2 (high rainfall)

Effect of Altering the Kd values

In this case the model was run with conservative Kd values. The conservative values were chosen assuming stronger association of the element with the soil solid phase, but still aimed to be reasonably realistic. As a consequence the extent of change in the Kd value was not the same for all elements. For example the Kd for thorium remained constant because under normal conditions it is already assumed to be very strongly sorbed to the soil solid phase. In contrast, for technetium a 'realistically' conservative value of 0.5 $m^3\ kg^{-1}$ can be envisaged which is 500X greater than the Kd predicted for it in the base case where it is assumed to be essentially unsorbed.

Table 15: Comparison of Conservative and Base Case Kd Values.

Element	Kd Values ($m^3\ kg^{-1}$) Base Case	Conservative
Tc	0.001	0.50
Np	0.01	1.00
U	0.10	1.00
Th	10.00	10.00

The results are shown in the table below as the factor by which the soil activity differed from that in the base case.

Table 16: Results of Assuming a Conservative Kd Value.

Nuclide	Time (a) 10	100	1000	10000
Tc-99	7.5	104	451	500
Np-237	1.5	11	71	100
U-233	1.2	6.4	328	556
Th-229	1.1	3.3	285	487

Effect of Reducing the Compartment Volumes

The system was modelled in exactly the same manner as the base case except that the volume of the groundwater, soil and soilwater compartments was reduced (Figure 2). This simulated the smaller valley of Hellikon, also located in northern Switzerland. 1 Bq l^{-1} Np-237 and Tc-99 were input into the system as before and the factor by which the soil activity differed from the base case is shown in table 17.

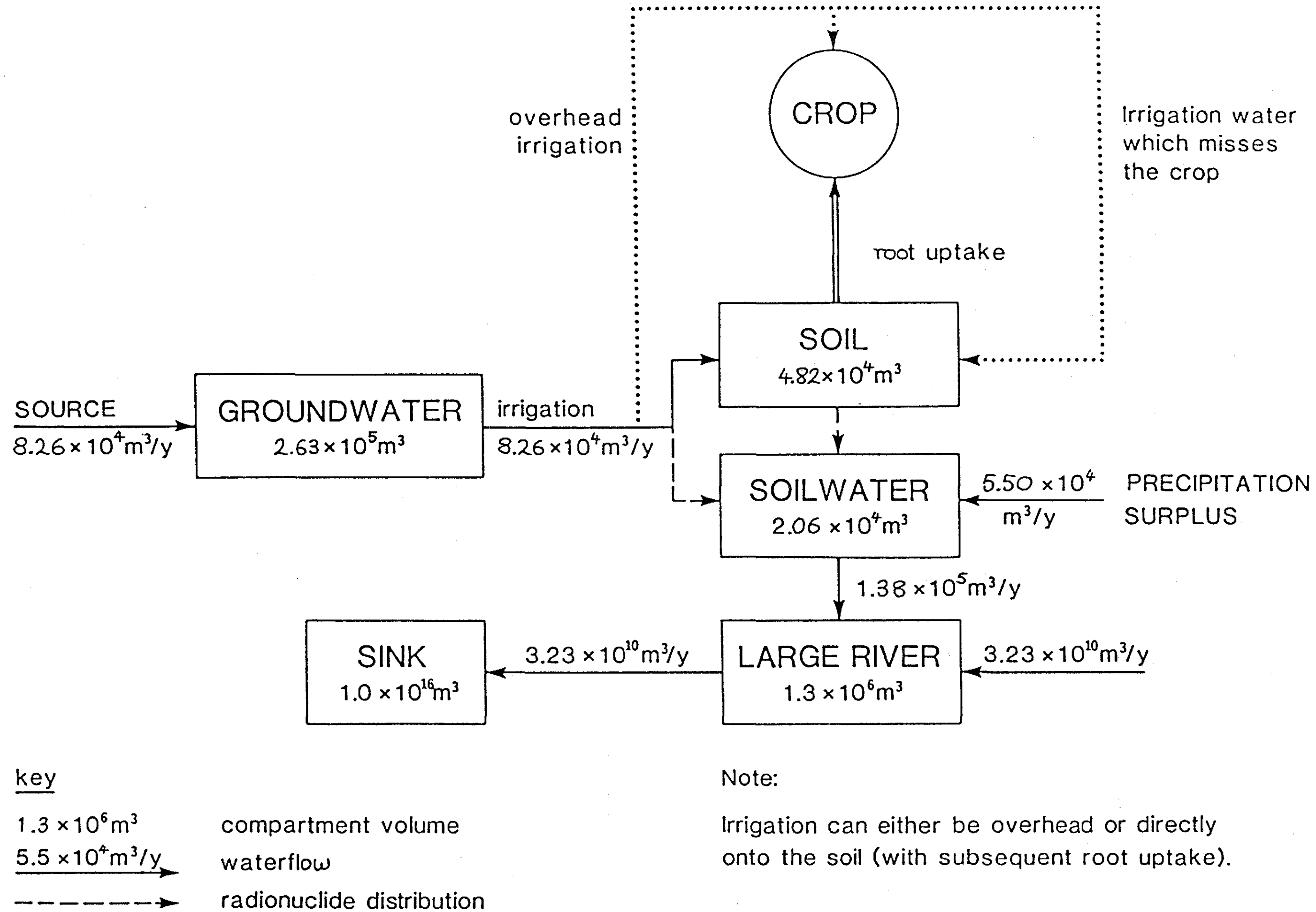


Fig. 2: Model with Reduced Compartment Volumes (Small Valley)

Table 17: Results of Reducing the Compartment Volumes.

Nuclide	Time (a)			
	10	100	1000	10000
Tc-99	1.30	1.0	1.0	1.0
Np-237	1.39	1.0	1.0	1.0
U-233	1.35	0.80	0.78	0.78
Th-229	1.44	0.79	0.77	0.80

Effect of Daughter Source Input

A futher variation assumed that the activity of all three radionuclides in the source term was 1 Bq l^{-1}. The table below shows the factor by which the calculated soil activity differed from the base case soil activity in which the source contained only Np.

Table 18: Result of Assuming Secular Equilibrium for Source Term.

Nuclide	Time (a)			
	10	100	1000	10000
Np-237	1.0	1.0	1.0	1.0
U-233	$4.6x10^4$	$1.7x10^4$	$1.6x10^4$	$1.6x10^4$
Th-229	$1.4x10^8$	$5.4x10^6$	$2.3x10^6$	$2.1x10^6$

6. Discussion

Uncertainty Estimates For Calculated Results

The uncertainty estimates were derived from subjective assessment of the parameter variation results and known variability of the input parameters.

Of the parameter variations studied it was found that increasing the rainfall has a diluting effect on radionuclide concentrations. A precipitation surplus of 200 mm a^{-1} (V2) [100% increase] resulted in the soil activities decreasing by up to 55% compared with the base case activities. Conversely when less rainfall is assumed (V1) [100% decrease] the resultant soil activities generally increase linearly, but in some cases (eg, Th-229 at very long times) this is closer to a factor three.

The effect of decreasing the compartment volumes (groundwater volume decreased by a factor of 24.7, soil/soilwater volumes decreased by a factor of 12.3) is to increase the radionuclide concentrations in the soil by up to 45% at short time intervals (less than 100 years). This is because the same volume of contaminated irrigation water is now passing through a smaller volume of soil so that the radionuclides accumulate more rapidly. However, steady - state is established at the same activity as in the base case for the parent radionuclides. In contrast, the daughter radionuclides establish equilibrium at a lower activity than in the base case ($\sim$ 20% less). This is a result of their relatively long half lives in comparison with the short turnover times for the compartments, whereby the radionuclides are removed before secular equilibrium can be established.

In contrast, the calculated activities were found to be highly sensitive to the Kd parameter. When conservative Kd values were input into the model (i.e., assuming stronger association of the radionuclide with the soil solid phase) the resultant activities increased by up to a factor of 560. The changes in calculated activities were most pronounced for the longer time periods (1000, 10000 years).

From these results it was concluded that over the long time periods (1000, 100000 years) the results of the Kd parameter variation could be judged to be maximum values for the calculated activities.

At the shorter time periods (10, 100 years) it was considered that the uncertainties in the soil-plant concentration ratios contributed most to the uncertainties in the final values (these uncertainties also hold for the long time periods). The compartment sizes and the rainfall regime were not considered to contribute a major source of uncertainty to the final calculated activities since they only exerted a two or three fold influence on the calculated activities.

It must be borne in mind that this is a subjective assessment of the uncertainties associated with a result and is in no way comprehensive. Not all parameters were considered although uncertainty in their values certainly exist. For example, in modelling overhead irrigation a lack of relevant experimental data prevented even a subjective assessment of the uncertainty associated with this calculation.

For all the calculations, the system was kept constant with time and it should be stressed that larger uncertainties are surely to be expected in the system with time. In addition to this, uncertainties in the system itself, such as how accurately reality has been described in the model, were not considered.

Special Remarks on the Decay Series

The radionuclide concentration in each compartment is very much determined by the physical rate of supply and removal of the radionuclides to each compartment. In the base case region modelled, the turnover times are all relatively short (e.g. groundwater 6.5 years), so that both Np-237 and Tc-99 attain steady - state in all the compartments in roughly 90 years. The Np-237 daughters also attain steady - state in the groundwater in less than 100 years but require much longer to establish steady - state in the soil/soilwater compartments (U-233 ∿865 years, Th-229 ∿100,000 years). This is a consequence of their high retention (Kd value) within the soil.

When the compartment turnover times are decreased further (groundwater 3.2 years) as a result of reducing the compartment volumes, the parent radionuclides accumulate more rapidly within each compartment although still attaining the same steady - state concentration as in the base case. This is because the radionuclides are transported more rapidly through the system thus accumulating faster, but since the source term concentration remains constant (1 Bq l^{-1}) identical steady - state concentrations result. The daughter radionuclides also accumulate faster in each compartment as they are also transported more rapidly through the system than in the base case. In contrast the steady - state concentrations are less than those in the base case because a given quantity of the parent radionuclide, which comprises the source term for the daughter, now passes through the compartment in a shorter time. Since a given quantity of parent requires less time to pass through the compartment, less time is available for the daughter's production. This process dominates because the daughter's half - life is very much longer than its residence time within the compartment.

The steady-state concentrations as obtained using BIOPATH can also be checked analytically. In the appendix the method of solving these analytical solutions is shown. When the transfer coefficients and decay constants are compared, it is clear from these solutions, that for a long-lived radionuclide the transfer coefficients are the determining factors for the steady-state radionuclide concentration.

When the input source term is set at 1 Bq l^{-1} for Np-237, U-233 and Th-229 (see table 18), very much higher concentrations of the U-233 and Th-229 result in the biosphere than when only the parent (Np-237) enters the system. Naturally this is because the daughter radionuclides each have a constant independent source term and are not relying solely upon the rate and extent of supply of the parent for their production.

For the BIOMOVS study the input source term was specifically defined as 1 Bq l^{-1} of the parent radionuclide Np-237 alone, however in terms of a safety repository assessment (such as Projekt Gewaehr) the daughter radionuclide is more likely to enter the biosphere already in secular equilibrium with its parent.

Irrigation Method

The relative importance of the two irrigation methods as a source of crop contamination varies with time. This difference in pathways is most marked for daughter radionuclides and radionuclides with a high soil Kd value. At short time intervals the overhead irrigation tends to dominate because the radionuclides rapidly attain steady - state in the groundwater used for irrigation, whereas the radionuclide activity in the soil has not yet accumulated to any appreciable extent. After a longer time period however, the groundwater concentration remains the same as before whereas the soil activity has had sufficient time to accumulate. This soil concentration may now be a more significant source of radionuclides (root uptake) than the overhead irrigation.

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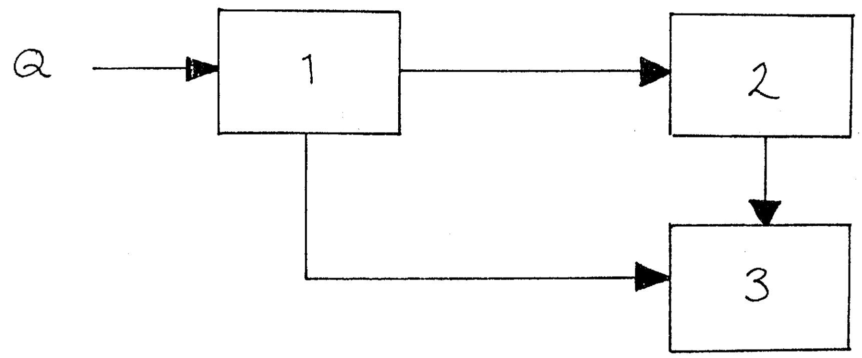
9. APPENDIX

Analytical Solutions For Steady-State In A Three Member Compartment Model

Model



The analytical solution for the steady - state concentration of a parent and daughter radionuclide in the compartments is given. For simplicity three compartments and two radionuclides in a decay chain are considered but this can be extended in principal to as many compartments and radionuclides as one desires.

Using the following general notation :-

Q [Bq/l] = source term concentration for radionuclide i in compartment k

i = radionuclide index for a decay chain $1 \rightarrow 2 \rightarrow 3 \ldots i-1 \rightarrow i \rightarrow \ldots$

C [mol/l] = concentration of radionuclide i in compartment k

$K_i^{j,k} [s^{-1}]$ = transfer coefficient for radionuclide i from compartment j to compartment k

λ_i $[s^{-1}]$ = decay constant for radionuclide i

we get the following balance equations.

Compartment 1

For the parent:-

$$0 = -\lambda_{i-1} C_{i-1}^1 + Q_{i-1} - K_{i-1}^{1,2} C_{i-1}^1 - K_{i-1}^{1,3} C_{i-1}^1$$

For the daughter:-

$$0 = -\lambda_i C_i^1 + Q_i + \lambda_{i-1} C_{i-1}^1 - K_i^{1,2} C_i^1 - K_i^{1,3} C_i^1$$

Compartment 2

For the parent:- $0 = -\lambda_{i-1} C^2_{i-1} - K^{2,3}_{i-1} C^2_{i-1} + K^{1,2}_{i-1} C^1_{i-1}$

For the daughter:- $0 = -\lambda_i C^2_i + \lambda_{i-1} C^2_{i-1} - K^{2,3}_i C^2_i + K^{1,2}_i C^1_i$

Compartment 3

For the parent:- $0 = -\lambda_{i-1} C^3_{i-1} + K^{1,3}_{i-1} C^1_{i-1} + K^{2,3}_{i-1} C^2_{i-1}$

For the daughter:- $0 = -\lambda_i C^3_i + \lambda_{i-1} C^3_{i-1} + K^{1,3}_i C^1_i + K^{2,3}_i C^2_i$

Analytical Solutions

Compartment 1.

$$C^1_{i-1} = \frac{Q_{i-1}}{(\lambda_{i-1} + K^{1,2}_{i-1} + K^{1,3}_{i-1})}$$

$$C^1_i = \frac{Q_i + \lambda_{i-1} C^1_{i-1}}{(\lambda_i + K^{1,2}_i + K^{1,3}_i)}$$

Compartment 2.

$$C^2_{i-1} = \frac{K^{1,2}_{i-1} C^1_{i-1}}{(\lambda_{i-1} + K^{2,3}_{i-1})}$$

$$C^2_i = \frac{(\lambda_{i-1} C^2_{i-1} + K^{1,2}_i C^1_i)}{(\lambda_i + K^{2,3}_i)}$$

Compartment 3.

$$C^3_{i-1} = \frac{1}{\lambda_{i-1}} (K^{1,3}_{i-1} C^1_{i-1} + K^{2,3}_{i-1} C^2_{i-1})$$

$$C^3_i = \frac{1}{\lambda_i} (\lambda_{i-1} C^3_{i-1} + K^{1,3}_i C^1_i + K^{2,3}_i C^2_i)$$