

# Individual radiation doses from nuclides contained in a WP-Cave repository for spent fuel

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**SVENSK KÄRNBRÄNSLEHANTERING AB** SWEDISH NUCLEAR FUEL AND WASTE MANAGEMENT CO

BOX 5864 S-102 48 STOCKHOLM TEL 08-665 28 00 TELEX 13108-SKB INDIVIDUAL RADIATION DOSES FROM NUCLIDES CONTAINED IN A WP-CAVE REPOSITORY FOR SPENT FUEL

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This report concerns a study which was conducted for SKB. The conclusions and viewpoints presented in the report are those of the author(s) and do not necessarily coincide with those of the client.

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#### INDIVIDUAL RADIATION DOSES FROM NUCLIDES CONTAINED IN A WP-CAVE REPOSITORY FOR SPENT FUEL

# Abstract

The individual radiation doses to man were calculated from leakage of radionuclides from a WP-cave repository for spent nuclear fuel. This study is a part of a safety analysis of a WP-cave repository performed by SKB.

It was assumed that the nuclides reach the biosphere by inflow to a well and/or a lake. Therefore calculations were performed for three different critical groups. The turnover in the biosphere and the exposure to man was modelled by a compartment model using the BIOPATH-code. The nuclides dominating the total dose were identified. These were C-14, Se-79, Sn-126, I-129, Pa-231, Th-229 and Np-237. Maximum doses were about 3E-6 Sv/year.

Uncertainty analysis was carried out using the PRISM-code for the dose dominant nuclides. For all nuclides and cases the ranges of the doses are within three orders of magnitude for 90 % confidence interval. The main parameters to the uncertainty in the case with outflow to a well are the volume of the well, the amount of water consumed or the migration in the soil. For the lake case major contributions to the uncertainty arise from the sedimentation, bioaccumulation in fish and the amount of consumption of fish.

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A	BIOPATH and PRISM codes
В	Reservoir masses and transfer coefficients
с	Input data for dose calculations

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#### 1 Introduction

A safety analysis for the WP-cave concept for storing high-level waste is at present being performed as a part of the SKB research program. This report handles the radiological consequences to man from long-lived nuclides contained in such a WP-cave repository. It is based upon two technical notes within the project (Bergström et al 1987, Bergström et al 1988)

The objective of the work described in this report is threefold:

- To present conversion factors between unit releases and doses to a specific ecosystem for all nuclides of radiological importance.
- To estimate doses to critical groups from calculated releases to the biosphere from actinide decay chains.
- To show the uncertainty in the doses predicted due to the uncertainty in the parameter values for the dose dominant nuclides.

The code BIOPATH (U Bergström et al, 1982) was used for calculations of the turnover of radionuclides in the biosphere and the resulting doses to man. The uncertainty analysis was carried out by using the PRISM-code (R Gardner et al, 1983).

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#### Background

Radioactive nuclides from a repository for spent fuel may reach the biosphere by groundwater transport. This contaminated water can expose man directly if it is utilized as drinking water. The turnover in the biosphere of the nuclides can also lead to accumulation in different food chains resulting in exposure to man. Accumulation in different compartments such as soil may also cause external exposure to man.

In this study it was assumed that the contaminated ground water reaches the biosphere through a well and/or outflow to a lake. The results are discussed in terms of maximum individual doses for three different hypothetical critical groups.

The considered recipient area is representative for the middle part of Sweden and mainly consists of wood lands with a minor part of farming land.

Because of the long time span involved for potential leakages of radionuclides the biosphere will undergo changes. These long-term changes are not possible to predict with accuracy. However representative scenarios reflecting the current state of the biosphere may be used for illustrating the possible consequences to man. In addition scenarios could be set up for different climate and stages of the society. In this report the doses were estimated from the present ecosystem through out the release period. The lake used in these calculations is not supposed to last for the time period studied but represents the consequences if a lake of this type is recipient anytime during the leakage. Doses from the nuclides in Table 2.1 were assessed.

Table 2.1

Nuclides, half-lives and dose factors (Sv/Bq) for intake via inhalation and ingestion. The actinides are sorted after respective decay chain.

Nuclide	Half-life Y=Years D=Days	Inhalation	Ingestion
C-14	5.7E3 Y	5.7E-10	5.7E-10
Ni-59	7.5E4 Y	3.6E-10	5.4E-11
Se-79	6.4E4 Y	2.4E-9	2.3E-9
Zr-93	1.5E6 Y	8.6E-8	4.2E-10
Nb-94	2.0E4 Y	9.0E-8	1.4E-9
Tc-99	2.1E5 Y	2.0E-9	3.4E-10
Pd-107	6.5E6 Y	3.4E-9	3.7E-11
Sn-121m	55 Y	2.5E-9	3.7E-10
Sn-126	1.0E5 Y	2.3E-8	4.7E-9
I-129	1.6E7 Y	4.7E-8	9.8E-8
Cs-135	2.3E6 Y	1.0E-9	1.9E-9
Np-237	2.1E6 Y	1.3E-4	1.2E-6
U-233	1.6E5 Y	3.6E-5	3.1E-7
Th-229	7 340 Y	5.7E-4	9.4E-7
Ra-225	14.8 D	2.0E-6	3.1E-7
Pu-239	2.4E4 Y	1.4E-4	1.2E-6
U-235	2.0E8 Y	3.3E-5	2.8E-7
Pa-231	3.2E4 Y	3.4E-4	2.2E-5
Ac-227	21.8 Y	1.0E-3	3.8E-6
Pu-240	6537 Y	1.4E-4	1.2E-6
U-236	2.3E7 Y	3.4E-5	2.9E-7
Th-232	1.4E10 Y	4.4E-4	7.4E-7
Ra-228	5.75 Y	1.2E-6	3.3E-7
Th-228	1.9 Y	8.8E-5	1.0E-7
Pu-242	3.8E5 Y	1.3E-4	1.1E-6
U-238	4.5E9 Y	3.2E-5	2.7E-7
U-234	2.5E4 Y	3.6E-5	3.0E-7
Th-230	7.7E4 Y	8.6E-5	1.6E-7
Ra-226	1602 Y	2.1E-6	3.1E-7
Pb-210	22.3 Y	3.4E-6	1.4E-6

References

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The calculations were performed for unit releases of the nuclides given in Table 2.1. These nuclides may reach the biosphere from a repository dependent on their long half-life or by generation from parent-nuclide.

For the decay chains, the doses were also calculated from the releases to the biosphere calculated by Moreno et al (1988). The source terms are only estimated for nuclides with longer half-lives than 1000 years. The daughter nuclides with shorter half-lives are only generated by the decay of respective parent nuclide in the biosphere.

Because models are simplifications of our perception of reality their results are affected by an inherent uncertainty. This uncertainty originates from different phases in the modelling and can be divided into the following head lines:

- interpretation of the scenario
- model structure
- parameter values
- human factors

The emphasis in this work was to examine the uncertainty in the results due to the uncertainty in the parameter values, for those nuclides which dominate the exposure.

#### 3 Description of codes

The mathematical method included in the BIOPATH code is based on compartment theory with firstorder kinetics. Therefore, the cycling and content of radioactive matter in different ecosystems are described by a system of first-order linear differential equations with constant or time varying transfer coefficients and a number of physically defined areas or volumes (which in this report are called reservoirs or compartments).

An integrated system of computer programs, PRISM was used for the uncertainty analysis. Within the PRISM-code, sets of model parameters are generated from given distributions of each parameter. The responses of the model are calculated for each set of those parameters and the results are analysed statistically. For description of the codes see Appendix A. 4

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#### Model of ecosystem

#### 4.1 Description of ecosystem

The radionuclides reach the recipient area via the transport with groundwater, into a well and/or into a lake.

The region represents a typical landscape from the middle part of Sweden. The area is mostly woodlands containing small agricultural areas.

Because of these condiditons the lake considered has an oligotrophic profile implying that the uptake of radionuclides by fish may be higher than that of a lake with a eutrophical profile. This is due to the lower contents of nutrients and ions of stable elements in an oligotrophical lake which results in a relatively higher accumulation to biota of the radionuclides. So is the case for cesium (Kohlemainen et al, 1966). However for actinides this information is often not available.

The lake in this study has a volume of  $3.2 \cdot E6 \text{ m}^3$ and a mean depth of 8 m. The residence time of water is about 3 years, because the drainage area to the lake is about 4.4 km<sup>2</sup> with an average runoff of 6.5 l/s, km<sup>2</sup> (Sundblad B et al, 1982).

#### 4.2 <u>Compartment structure</u>

Application of the BIOPATH-code implies that according to the compartment theory the biosphere to be studied is divided into appropriate reservoirs or compartments. The number and structure of compartments represent a compromise between:

- a sufficiently differentiated system in order to encompass all important reservoirs and exposure pathways

- simplicity of design in order to facilitate uncertainty analyses and comparison of model predictions with measurements of turnover and elemental balance in nature or calculations using other models, and
- available information on dispersal mechanisms.

In general compartments can be designed so that they with satisfactory precision fulfill the condition of momentaneous homogeneous mixing. In nature, however, such reservoirs are often connected to areas with gradients or where the probability of leaving the reservoir may vary substantially within the reservoir, like the sediments. Better realism is then obtained by further division of such reservoirs.

Ten reservoirs were designed for simulation of the turnover of radionuclides in this ecosystem. A schematic description of the compartment system is given in Fig 3.1. The contaminated ground water enters the system in a water reservoir representing the well. From this reservoir water is taken for irrigation of a small garden plot. The soil constituting this garden plot is the next reservoir in the system. Nuclides migrate in the soil and will with time reach the deeper part of the soil however; with different rate due to their physical and chemical properties. A deeper soil reservoir connected to the upper one is used in order to simulate this process.

The migration of the radionuclides may also lead to transport back to the ground water. Minor amounts of the radionuclides can thus reach the well

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once again. This circulation, though of minor importance for the doses, is simulated in the model by the transport from local deep soil back to the well.

The water passing the well flows into a lake, which is symbolized by one reservoir for the water. This is of course a simplification due to inhomogeneous mixing especially during time periods when there is a stratification in the water column. However, for annual average conditions this is an acceptable generalization.

The sediments in the lake is represented by two reservoirs physico-chemical and biological, one upper aerated zon where bioturbation mostly occurs and one deeper with reducing conditions acting in this case as a sink for the nuclides.

Further, it is assumed that the water in this lake is used for irrigation of farming land in the vicinity of the lake. This area constitutes of three reservoirs describing the plough-layer, deeper soil and groundwater respectively. This top soil consists of the soil which is irrigated with water from the lake. The nuclides migrate to deeper layer and back to the lake. In this case this recirculation is described with two compartments, deep soil and ground water.

Finally there is one atmospheric reservoir for the air overlying the ecosystem. In the figure describing the system the masses are also given. As can be seen the volume of the well was 250 000 m<sup>3</sup> which is the same value as used in the KBS 3 study (Bergström 1983). In addition, a much lower volume 2 000 m<sup>3</sup> was also adopted for dose-dominating nuclides.

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These volumes represent the total volume of water available for dilution of the radioactive nuclides from the repository to the entrance into the biosphere.



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## Figure 4.1

The structure of the compartment model with the masses (kg).

- \* Calculations performed with two volumes of the well.
- + Not of importance for the results.

### 4.3 Transfer processes

The transfer processes considered are mathematically described as transfer coefficients (turnover per year) in the code. There are two groups of transfer coefficients namely the general and the element dependent. The general coefficients are depending on physical processes only, while the element dependent also are due to chemical processes. For the former, information about the turnover of different carriers in each media may be used for obtaining the necessary coefficients, like the turnover of water in the lake or the amount of water used for irrigation. For the latter additional information about the solubility of each element must be considered. In general this is described by a distribution factor  $K_d$ .

It was assumed that the nuclides are in soluble form in the well water with no interaction with particles.

Nuclides reaching the well are mainly carried out from the well by the turnover rate of water. This turnover rate is assumed to be twice a year for the large well and once a year for the small one.

Furthermore nuclides reaching the well are carried to the local top soil by using water from the well for irrigation of the garden plot. This is described by a general transfer coefficient (k) in the following way:

$$k = A/W (y^{-1})$$

where

A = The volume of water for irrigation of 400 m<sup>2</sup> per year (m<sup>3</sup>/y) with 50 mm per m<sup>2</sup>.

W = Volume of the water reservoir (m<sup>3</sup>).

The nuclides migrate in the soil, the result of which is a transport to deeper layers and to the ground water. This transport is dependent on environmental conditions as well as on the physical and chemical behaviour of the radionuclides. In Appendix A the equations and data used for obtaining these transfer coefficients are given, as well as the values for the transfer coefficients.

When radionuclides reach the water in the lake depletion of the nuclides in the water will occur due to interaction with mineral and organic particles causing a transport to the sediment. This process is dependent on the chemical form of the radionuclides and the environmental conditions. In Appendix B the equation used, the parameter values and resulting transfer coefficients are given for the transfer from water to the sediment of each element.

Several processes like bioturbation, diffusion and convection may affect the radionuclide transport within the sediments and may cause resuspension of radionuclides from the sediment surface back to the water. It is well recognized that there is a lack of data to describe this process in detail. However, in a report handling estimation of major uncertainties coupled to modelling of the turnover of Cs-137 and Ra-226 in a lake ecosystem the uncertainty in the transfer had a low impact on the resulting doses (Bergström et al, 1984). If the transport back to the water from the sediment is mostly cused by a mecanical process the transfer rate will be the same for all nuclides. This was assumed in this study. For the calculations presented below a value of 0.001 was chosen, which is in the same order of magnitude as values reported in BIOMOVS 1988. However, in shallow lakes this

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value can be considerably higher causing a redistribution of the activity within the lake sediments.

Nuclides in the upper sediment are also transported to deeper sediments due to the annual growth of the sediments, diffusion and other processes. Assuming that the annual growth of sediments is the main process and that the total mass of the top sediments is constant, a general transfer coefficient can be calculated. This growth is about 1 kg/m<sup>2</sup>y leading to a transfer coefficient of 0.03 y<sup>-1</sup> for all nuclides.

Nuclides in the water column are also transferred to the soil by using water in the lake for irrigation of farmland with 150 mm per m<sup>2</sup> and year. For obtaining the transfer coefficient an assumption of the size of irrigated area was a necessity, though the most important feature is how much water is given per unit area. Naturally the irrigated area must be sufficient for producing the annual foodstuff necessary for critical group.

Nuclides reaching the soil migrate in a similar way with the process described above, see appendix B. Reaching the goundwater underlying the irrigated soil there is a feedback to the lake by the groundwater discharge to the lake.

There is also a transport to the atmosphere mostly caused by erosion and resuspension. This is a general process independant of the element to be studied. Recycling back to the soil occurs by deposition from the atmosphere. To summarize there are several processes and recycling encountered in the model, each described by a transfer coefficient, which of course has different impacts on the doses obtained.

#### 4.4 <u>Concentration and distribution factors</u>

The uptake of radioactive nuclides in biota from the surrounding media, soil and water etc are dependent on many factors. For example, the uptake in plants can be quite different from sandy and clayey soil and is also dependent on the pH in the soil. For essential elements the uptake in fish may differ due to trophical level of the fish and type of lake. In general, the uptake is higher for oligotrophic lakes than for eutrophic lakes.

This uptake is for steady-state conditions expressed as a bioaccumulation factor or concentration factor. It is an empirical ratio of nuclide concentration in the tissue of an organism to that in the connected media such as water or soil.

The concentration factors found in the literature are rather general, although in some cases they are related to special conditions in the environment or results from specific experiments. All the previously mentioned factors introduce a large variation between data from different sources.

Despite all these variations a reasonable value for the concentration factors of the most important food items, as well as for the pasturage for cows can be given. The values used in this report are given in Appendix C, Table C.1, with brief comments about how each value was selected. In Table C.2 the ranges are given for those nuclides which were included in the uncertainty analysis.

In order to make a conservative estimate the highest value for the concentration factors was taken when e.g. the uptake from different types of soil was found in the literature.

To describe the transfer from animal feed to animal products a distribution factor is used. This factor gives the fraction of the daily intake of nuclide which at steady state is likely to be found per litre of milk or per kilo of beef. The distribution factors used in this report are given in Appendix C, Table C.3 and the ranges in Table C.4.

# 4.5 <u>Exposure pathways</u>

Man receives radiation doses via different paths of exposure. Earlier calculations of the radioactive nuclides transport through the different reservoirs in models of the biosphere, showed that the internal exposure through inhalation, consumption of food-stuff and drinking water dominates the exposure for most of the nuclides. The pathway for inhalation is through inhalation of dust from the irrigated soil. Internal exposure from food can take place via a number of links in the ecological transport chain such as

- uptake in crops via root uptake
- uptake via the food chain of grass-meat,
  grass-milk and grain-egg
- uptake in fish or other marine foodstuffs from surrounding water

External radiation from ground contaminated by irrigation is also included.

According to the radiological definition, the critical group shall consist of a limited number of individuals who can receive higher doses than average. Doses to critical groups were calculated for three different exposure scenarios.

#### Case 1 (well scenario):

The critical group is exposed through the activity in groundwater representing the well (two different dilution volumes). The water in this well is used for the drinking demands of people and cattle. In addition, a garden plot is irrigated with water from the well. This gardenplot produces the annual demands of vegetables and root vegetables.

#### Case 2: (lake scenario)

The critical group is exposed to the activity reaching the lake. Drinking water for man and cattle is taken from the lake. All terrestrial products consumed are taken from the soil contaminated by irrigation from the lake. Exposure from consumption of fish from the lake is also considered.

# Case 3 (mixed scenario):

The critical group is exposed from nuclides both in the groundwater (well) and the lake. The case represents a situation with a well with outflow to a lake. This group uses water from the well for consumption of water by man and cattle and for irrigation of a garden plot where vegetables and root vegetables grow for consumption. Their annual amount of cereals is taken from the soil contaminated by irrigation from the lake. The fish consumed is contaminated by the activity in the lake.

A schematic presentation of how the exposure pathways are calculated for each case is shown in Figures 4.2 - 4.4. A summary of pathways considered for the different critical groups is given in Table 4.1.

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Consumption data used are given in Appendix C. Note that the "low volume" well has decreased consumption values due to decreased production as result of decreased irrigation. The yield can not cover the demand for the critical group. The values are shown in Table C.6.

#### Table 4.1

Exposure pathways for the critical group.

	Case 1	Case 2	Case 3
Inhalation	well	lake	lake
Drinking-water	well	lake	well
Milk and meat	well	lake	well/lake
Meat	well	lake	well/lake
Cereals	nc	lake	lake
Leafy vegetables	well	lake	well
Root vegetables	well	lake	well
Egg	well	lake	well/lake
Fish	nc	lake	lake
Ground cont	well	lake	lake

nc = not considered

Although, milk, meat and eggs are considered as exposure pathway for all three different hypothetical critical groups, the paths of intake of activity by the animals vary. In Case 1 the only intake by cow and hen is by drinking-water. In Cases 2 and 3 additional pathways are considered such as root-uptake, retention on the surfaces of vegetation from irrigation and deposition as well as consumption of soil when grazing. However, in Case 3 (mixed case) drinking-water for animals is taken from the well.



#### Figure 4.2

Exposure pathways for the critical group in case 1 (well scenario).

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Figure 4.3

Exposure pathways for the critical group in case 2 (lake scenario)

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# Figure 4.4

Exposure pathways for the critical group in case 3 (mixed scenario).

# 4.6 <u>Dose factors</u>

In order to transform the intake of 1 Bq of a nuclide to radiation dose, a dose conversion factor is used. This factor is dependent on the exposure situation, the decay energy of the nuclide, type of radiation and turnover in the body, and is based on data and calculations taken from ICRP.

The dose conversion factors used are the sum of ICRP's weighted organ-dose equivalent commitments, according to ICRP30, except for Pu- and Np-isotopes where the values from ICRP48 are used. The dose factors for inhalation and ingestion are given in Table 2.1.

The external dose from top soil is calculated with the dose conversion factors from Svensson (1979) with assumption of homogeneous distribution of the activity in soil. The compartment of top soil represents a depth of 30 cm. The contribution to the external dose from activity in deeper soil is only a minor part due to the absorption of the radiation in the soil.

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### Releases to the biosphere

For each nuclide the doses from continuous leakages of 1 Bq/year were calculated including the contributions from daughter nuclides generated by decay in the biosphere. Furthermore, dose calculations were performed for calculated releases to the biosphere of the actinide chains U-238, Np-237, Pu-239 and Pu-240.

The source terms to the biosphere were for U-238 calculated with solubility limitation within the cave (K Skagius, 1989) leading to a lower release than if the solubility limitation had not been taken into account. The releases of each nuclide to the biosphere were presented by Moreno et al (1989).

The release of the actinides to the biosphere are shown in Figures 5.1 and 5.2.

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# Figure 5.2

Release to the biosphere of the U-238 chain considering solubility limitations.

#### Results

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The results are discussed in terms of maximum annual doses representing steady state conditions in the biosphere. If there are radioactive decay products the contribution from them are included in the doses. For the estimated releases of actinides the dose from each nuclide is given as well as the total dose from the chain.

Comments and detailed tables of results are given below for each exposure scenario and the two types of source terms considered.

However, the dose calculations for C-14 were carried out by a specific activity method due to the behaviour of carbon in nature. Carbon is one of the major elements in all living organisms. There is almost no difference between the behaviour of stable or radioactive carbon. For a lake case the dominant exposure pathway is through consumption of fish (Bergström U et al, 1987). At steady state the time for turnover of C-14 is equal to that of the stable carbon. With the volume and turnover rate of water in this lake the concentration in the water becomes 1 nBq per 1 per unit release. In an oligotrophic lake the concentration of carbon in the water is approximately 10 ppm and the carbon content in fish is 0.005 kg per kg of fish. At steady state in the system this results in a C-14 content in fish of 5 µBq per kg. With an annual consumption of 30 kg, the dose becomes 9 E-14 Sv/year. A judgement is that this pathway contributes with up to 99 percent of the total dose.

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## 6.1 <u>Unit releases</u>

The results are presented in Tables 6.1 to 6.3 for the three different cases respectively. In addition the obtained doses from the extreme well case are given in Table 6.4. In this case the calculation was only performed for a selection of nuclides. Comments on the results are given below for each critical group (exposure scenario).

#### Case 1: Well

Doses are presented in Tables 6.1 and 6.4 for the base case well and the low volume well, respectively. In the latter table, a factor is also given describing the increase of doses when the total available dilution volume is decreased.

The dominant exposure pathway is via consumption of water for almost all nuclides. For most of the activation and fission products the additional contributions of importance are via consumption of vegetables and/or milk. Only for Nb-94 the main pathway is via external exposure from ground.

The relative contribution of the drinking water pathway increased somewhat for the "small" well due to the decreased amount of water from the well used for irrigation. For this latter case a decreased production and consumption of vegetables were assumed.

#### Case 2: Lake

Results are presented in Table 6.2. The main pathway for most of the nuclides in this case is via consumption of fish.

For Pu-isotopes and Th-isotopes the exposure is dominated by inhalation because of their high dose conversion factor for inhalation and their low mobility in soil compared to the other nuclides. However, these inhalation doses are conservatively biased because of the continuous irrigation until steady state conditions are reached.

#### Case 3: Mixed

Results are presented in Table 6.3. For all actinides the exposure is dominated by the exposure from drinking-water, taken from the well. However for most fission and activation products the fish pathway dominates the dose, because of their generally higher uptake to fish-flesh compared to the uptake of actinides. Similarly to case 1, the exposure from Nb-94 is dominated by external exposure.

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# Table 6.1

Case 1 A (well scenario) Individual doses from unit releases. The actinides are sorted after respective decay chain.

Nuclide	Dose	Dose from parentnuclide	Dose via consumption
	Sv/year	90	%
C-14 Ni-59 Se-79* Zr-93*	5.0E-16 6.6E-17 6.8E-15 2.4E-15	- - 56	99 72 30 15
Nb-94** Tc-99 Pd-107 Sn-121m Sn-126	2.3E-13 3.1E-16 4.4E-17 4.5E-15 6.0E-15	- - 78 84	1 95 74 83 73
I-129* Cs-135*	2.7E-13 3.2E-15	-	32 46
Np-237 U-233 Th-229 Ra-225	1.2E-12 3.1E-13 1.7E-12 3.2E-14	99.9 99.4 70	90 89 64 90
Pu-239 U-235 Pa-231 Ac-227	1.4E-12 2.8E-13 2.3E-11 3.7E-12	100 98.8 94 99.8	80 90 86 89
Pu-240 U-236 Th-232 Ra-228 Th-228	1.2E-12 2.9E-13 1.4E-12 3.3E-13 7.5E-14	100 100 75 93	83 90 49 86 99
Pu-242 U-238 U-234 Th-230 Ra-226 Pb-210	1.4E-12 2.7E-13 3.0E-13 2.3E-13 3.4E-13 1.2E-12	100 98 100 92 -	71 85 87 67 84 98

\* Food: Se-79 70 %, I-129 69 %, Cs-135 54 % \*\* External 98 %

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# Table 6.2

Case 2 (Lake scenario) Individual doses from unit releases. The actinides are sorted after respective decay chain.

Nuclide	Dose	Dose from parent- nuclide	Dose from intake of water	Dose from intake of fish
	Sv/year	ð 	<u>у</u> б	50 
C-14	8.6E-14	-	0	99
Ni-59	4.3E-17	-	12	81
Se-79	4.9E-14	-	1	97
Zr-93	1.1E-15	69	6	23
Nb-94* Tc-99 Pd-107 Sn-121m	8.2E-15 3.2E-16 2.9E-17 2.4E-15	- - 78	1 46 12 0	0 47 83 99
Sn-126	2.5E-14	97	0	99
I-129	3.3E-13	-	12	42
Cs-135	1.2E-13	-	0	99
Np-237	2.0E-13	99	57	39
U-233	1.4E-13	99	22	74
Th-229	3.8E-13	92	5	10
Ra-225	6.9E-16	-	35	59
Pu-239	3.3E-13	100	5	11
U-235	1.2E-13	99	22	74
Pa-231	2.1E-12	47	15	11
Ac-227	1.0E-12	99.8	32	55
Pu-240	2.3E-13	100	7	15
U-236 Th-232 Ra-228 Th-228	1.3E-13 4.2E-13 7.9E-14 3.5E-14	100 80 98	22 3 33 1	74 6 58 1
Pu-242	4.2E-13	100	4	7
U-238	1.2E-13	99	22	74
U-234	1.3E-13	100	22	74
Th-230	6.7E-14	98	4	7
Ra-226	9.0E-14	-	34	58
Pb-210	4.4E-12	-	13	87

\* External 98 %

,

# Table 6.3

Case 3 (mixed scenario) Individual doses from unit releases. The actinides are sorted after decay chain.

Nuclide	Dose	Dose from parent-	Dose from intake of	Dose from intake of
	Sv/year	% 		2 1 S I I
C-14 Ni-59 Se-79	8.6E-14 1.0E-16 5.5E-14	-	1 47 4	99 34 87
Zr-93 Nb-94* Tc-99 Pd-107	2.4E-15 1.2E-14 4.8E-16 6.8E-17	52 - - -	16 10 62 48	11 0 32 35
Sn-121m Sn-126 I-129 Cs-135	2.8E-15 3.0E-14 4.7E-13 1.3E-13	79 96 -	14 15 18 1	81 82 29 97
Np-237 U-233 Th-229 Ra-225	1.2E-12 4.1E-13 1.8E-12 3.3E-14	100 99.6 85	87 67 64 88	6 24 3 1
Pu-239 U-235 Pa-231 Ac-227	1.4E-12 3.7E-13 2.4E-11 4.3E-12	100 99 96 99.8	75 67 81 77	2 24 1 14
Pu-240 U-236 Th-232 Ra-228 Th-228	1.3E-12 3.8E-13 1.5E-12 3.7E-13 7.7E-14	100 100 72 96	83 67 61 77 96	3 24 2 12 3
Pu-242 U-238 U-234 Th-230 Ra-226 Pb-210	1.3E-12 3.6E-13 3.9E-13 2.5E-13 3.9E-13 5.1E-12	100 99 100 91 -	66 67 67 55 74 24	2 24 24 2 13 76

\* External 66 %

#### Table 6.4

Case 1B (well scenario) Individual doses from unit releases and relative increase of the doses compared to the base well (Case 1A).

Nuclide	Dose (Sv/year)	Dose from consumption of water	Relative increase
Se-79	7.0E-13	72	100
Sn-126	1.1E-12	91	180
I-129	3.7E-11	58	140
Pu-242	2.7E-10	90	190
Np-237	2.7E-10	96	230
U-236	7.2E-11	89	250
Pa-231	5.1E-09	95	220
Th-229*	3.1E-10	87	160
Ra-226	7.6E-11	88	220
Pb-210	3.0E-10	99	250

\* Including contribution from daughter nuclide Ra-225.

# 6.2 Estimated leakages of actinides

The results of each decay chain are presented in Figures 6.1 and 6.2 for the Case 1A and 2. respectively. Results for each nuclide and case are presented in Tables 6.5 - 6.7. No calculations were performed for the low-volume well. The Np-237 chain gives the highest contribution to the total dose, see Fig 6.1 and Fig 6.2, caused by the nuclides Th-229, Np-237 and U-233, (Tables 6.5 and 6.6). Thereafter, follows the Pu-239 chain of which it is Pa-231 that gives the highest doses. The leakage of the nuclides in the U-238 chain is calculated with solubility limitations within the cave (K Skagius, 1988). This effect if considered for the other chains would also reduce the leakage rates. In Tables 6.5 to 6.7 the dominant pathways are also given. They are likewise those pathways for the unit releases. Consumption of drinking-water

dominates the total exposure for Cases 1 and 3 respectively, while inhalation dominates for Case 2. This is because of the higher dilution volume in the lake water compared to the well and the high accumulation in soil. All actinides in general have low transfer to animal products like milk and meat. The root-uptake is also quite low for these nuclides. Unfortunately the data are often scarce and sometimes contradictory concerning the uptake to fish-flesh. However, the values used for the bioaccumulation factor to fish were mostly biased against the higher values in order not to underestimate this pathway and the doses.



Total dose from actinides with case 1 A (well scenario) with contribution from respective chain.



# Figure 6.2

Total dose from actinides Case 2 (lake scenario with contribution from respective chain.

Table 6.5

Case 1 A (well scenario). Individual doses from calculated releases to the biosphere for each chain and nuclide.

Nuclide	Dose (Sv/year)	Pathways, percent Inhalation	of total dose Drinking- water	from: Food
Np-237-c] Total Np-237	hain 3.1E-06 1.0E-06	5	76 91	18
U-233 Th-229 Ra-225	3.7E-07 1.2E-06 4.8E-07	16	87 69 49	13 14 50
Pu-239-ch Total Pu-239 U-235	nain 7.5E-07 1.3E-08 8.6E-09	3 15	80 75 87	16 8
Pa-231 Ac-227	7.0E-07 4.2E-08	1 46	84 4	15 15 51
Pu-240-ch Total Pu-240 U-236	nain 1.2E-07 1.2E-11 1.2E-07	9	87 83 87	12 4 12
Th-232 Ra-228 Th-228	4.4E-11 1.4E-11 2.4E-12	19 63	65 6 1	17 94 35
U-238-cha Total U-238 U-234 Th-230 Ra-226	in* 1.0E-09 3.3E-11 6.4E-11 2.2E-11 1.0E-09	17	85 87 87 64 83	15 12 12 17 16

\*

The release rate to the biosphere is calculated with regard to the solubility limited concentration.
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# Table 6.6

Case 2 (lake scenario). Individual doses from estimated release to the biosphere for respective chain and nuclide.

Nuclide	Dose	Pathways, p Inhalation	Dercent of Drinking	total dos - Food	e from: Fish
	(Sv/year	)	water		
Np-237-c	hain				
Total	7.2E-07	44	21	4	31
Np-237	1.7E-07		57	4	39
บ-233	1.6E-07	1	22	4	73
Th-229	3.4E-07	87	4	1	8
Ra-225	3.2E-08	5	19	46	32
Pu-239-cl	nain				
Total	6.5E-08	54	17	14	15
Pu-239	3.2E-09	84	5		11
U-235	3.7E-09		22	4	74
Pa-231	3.1E-08	22	31	27	21
Ac-227	3.2E-08	94	1	3	2
Pu-240-ch	nain				
Total	5.5E-08	1	21		73
Pu-240	2.3E-12	77	7		15
V-236	5.5E-08	1	21	4	73
Th-232	1.5E-11	89	3	1	, 5
Ra-228	1.2E-12	3	11	67	19
Th-228	2.5E-12	99		07	
U-238-cha	in*				
Total	2.9E-10		33	7	59
U-238	1.4E-11		22	4	74
U-234	2.8E-11	1	21	4	73
Th-230	6.5E-12	88		1	7
Ra-226	2.6E-10	1	32	11	ςς
		-		**	

\*

The release rate to the biosphere is calculated with regard to the solubility limited concentration.

# Table 6.7

Case 3 (mixed scenario). Individual doses from estimated release to the biosphere for each chain and nuclide

Nuclide	Dose	Pathways, percent of total d		tal dose	se from:	
	(Sv/year)	Innalation	Drinking- water	Food	Fish	Ground cont
Np-237-ch	ain					
Total Np-237 U-233	3.3E-06 1.1E-06 4.9E-07	9	68 85 65	16 9 10	7 6 24	
Th-229 Ra-225	1.3E-06 5.1E-07	24	63 48	12 49	2 2	
Pu-239-ch	ain					
Total Pu-239 U-235	7.6E-07 1.4E-08 1.1E-08	5 19	78 73 66	16 5 9	1 2 24	
Pa-231 Ac-227 Th-227	7.1E-07 5.4E-08 1.2E-10	1 55 56	84 3 3	15 43 37	1 1 1	3
Pu-240-ch	ain		-		-	5
Total Pu-240 U-236	1.6E-07 1.3E-11 1.6E-07	14	65 80 65	10 4 10	23 3 23	
Th-232 Ra-228 Th-228	5.0E-11 1.4E-11 3.3E-12	73	57 5 1	13 93 26	2 1	
U-238-chai	in*					
Total U-238 U-234	1.1E-09 4.4E-11 8.5E-11	1	72 65 65	14 10 10	13 23 23	
Th-230 Ra-226	2.3E-11 1.0E-09	24	60 71	14 14	2 13	

\*

The release rate to the biosphere is calculated with regard to the solubility limited concentration.

# 6.3 <u>Dose dominant nuclides</u>

By using the dose conversion factors for unit releases of fission and activation products and calculation of actual leakages to the biosphere the expected doses can be assessed. This is because the turnover rates of importance for the doses are higher than the duration of the maximum inflow to the biosphere. This approximation may be satisfactory due to the other sources of uncertainty. The results of this estimation for the activation and fission products are given in Table 6.8. The exposure is dominated by the contributions from C-14, Se-79, Sn-126 and I-129.

However, for decay chains the situation is more complicated which implies the necessity of carring out complete calculations for the calculated releases. For the actinides, see 6.2, the exposure in the well case is dominated by Np-237, Pa-231 and Th-229.

The highest doses for all nuclides are obtained for the mixed scenario. For all actinides the well scenario gives higher doses compared to the lake scenario. For the dose dominant activation and fission products the lake scenario is the most critical. This is because of the higher bioavailability for these nuclides, which results in a greater contribution from the food chains.

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# Table 6.8

Maximum doses for estimated releases of activations and fission products.

	Estimated		Dose (Sv/year)	)
Nuclide	Releases* (Bq/y)	Well Case	Lake Case	Mixed Case
C-14	4.6E7	2.3E-08	4.0E-06	4.0E-06
Ni-59	1.1E6	7.3E-11	4.7E-11	1.1E-10
Se-79	2.7E7	1.8E-07	1.3E-06	1.5E-06
Zr-93	3.4E4	8.2E-11	3.7E-11	8.2E-11
Nb-94	<1	-	-	-
Tc-99	9.2E4	2.9E-11	2.9E-11	4.4E-11
Pd-107	3.5E2	1.5E-14	1.0E-14	2.4E-14
Sn-121m	<1	-	-	-
Sn-126	9.6E7	5.8E-07	2.4E-06	2.9E-06
I-129	2.7E6	7.3E-07	8.9E-07	1.3E-06
Cs-135	3.4E5	1.1E-09	4.1E-08	4.4E-08

\* Ref Moreno et al 1989

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### Uncertainty analysis

For the dose dominating nuclides in the well scenario uncertainty analyses were performed for the well and lake cases respectively. These nuclides were Se-79, Sn-126, I-129, Np-237, Th-229 and Pa-231. C-14 was not included due to the special method of calculation. These cases were selected to clearly illustrate the uncertainty in the different pathways. In addition it is possible to draw conclusions for the mixed case from the combination of the cases studied.

# 7.1 <u>Parameter variation</u>

There are two main sources to the uncertainty in parameter values. The first is variation caused by the natural variability, while the next is due to incomplete knowledge or ignorance about the behaviour of nuclides released to the biosphere.

In the performed uncertainty analysis all parameters used to calculate the turnover in the biosphere and the exposure to critical group were varied. However, the masses of the reservoirs were not included with the exception of the volume of the well, describing the total dilution of contaminated groundwater reaching the biosphere. In addition the source term was kept constant.

The dilution volume for the well in the base case was varied considerably within 100 to 500 000 m<sup>3</sup>, assuming a log uniform distribution.

The values used in the BIOPATH-calculations are the best estimates for all data. In the uncertainty analysis all the data used to calculate the transfer coefficients for migration in soil

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and sedimentation were varied. These ranges are all given in Appendix C. How the consumption data were varied are given in Appendix B. Most of the parameters were assumed to belong to triangular distributions. The ranges were chosen to be within reasonable limits, though they may represent extreme consumption values, e.g. 880 1 of milk per year. However, all minimum consumption values were greater than zero. This was because the contribution from each pathway should always be considered. In some cases the best estimate is close to the maximum value. In these cases the values in the model calculations were chosen conservatively.

In the "small" well case some parameters were changed compared to the basic well case. These data are given in Appendix C Table 6.

How the element specific parameters were varied (type of distribution, best estimate values and ranges) is given in Appendix C. In general the ranges used were based on values found in the literature. However, when pertinent information was not available a factor of ten up and down from the best estimate value was used.

The importance of type of distribution used can not be generally treated as the outlook of the distribution randomized is not only depending on type chosen. The relation between the different input parameters such as ranges and most probable value is also of importance.

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# 7.2 Results from uncertainty analysis

Uncertainty analysis was performed for Case 1 (well scenario, two different volumes) and Case 2 (lake scenario).

The resulting ranges of the relative uncertainty are presented graphically in Figures 7.1 to 7.3. The ranges in the figures are the 5th and 95th percentile of the dose for each nuclide.

The ranges of uncertainty are within three orders of magnitude for most nuclides.

For all nuclides the ranges of uncertainty decrease considerably from Case 1 A to Case 1 B. This reflects directly the smaller range used for the dilution volume in the latter case compared to the former. Also the lake case had lower ranges of the uncertainty, though many parameter values varied within a factor of hundred. Below discussions of the results for each scenario are given.

The regression analysis showed high R2 values (regression sum of squares divided with total sum of squares) above 80, which indicates that conclusions can be drawn regarding which parameter dominated the uncertainty.



# Figure 7.1

Relative uncertainty for the well (Case 1a). Ranges within 5 to 95 percentiles.



# Figure 7.2

Relative uncertainty for the small well (Case 1b). Ranges within 5 to 95 percentiles.



### Figure 7.3

Relative uncertainty for the lake scenario (Case 2). Ranges within 5 to 95 percentiles.

Case 1A: well scenario

As can be seen from Fig 7.1 the ranges for all nuclides are large. This is due to the wide range of the dilution volume. For all nuclides the exposure from drinking water dominated the dose as can be seen in Fig 7.4. Consequently this pathway also dominates the uncertainty. For Se-79, Sn-126 and I-129 additional contributions come from consumption of meat and milk. The main parameters are shown in Fig 7.5. As expected the dilution volume of water is the parameter that have the highest correlation to the total dose, leading to the highest contribution to the uncertainty in total dose.



Figure 7.4

Relative contribution from dominant exposure pathways to total dose (D) and uncertainty (U) in the well scenario (Case 1A).





Dominant parameters to the uncertainty in the well scenario (Case 1A).

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### Case 1B: Small well scenario

In Fig 7.6 the relative contribution to the total dose and uncertainty for the dominating pathways are shown. In this case the ranges from the uncertainty analyses are much smaller due to the small range of the dilution volume. For most nuclides the main pathway still is via consumption of water but the other pathways give a greater contribution than in the base case. This is due to the assumption that a greater part of the total volume is used for irrigation leading to a relatively higher concentration in the soil for this case. This effect can especially be seen for Se-79 where the dominant pathway in the former case was via drinking water while in this case consumption of grown products from the garden plot is the main pathway. The same effect is also valid for Th-229, but here the pathway is inhalation of dust. The parameters describing the migration in soil will therefore give considerable contribution to the uncertainty for these two nuclides as can be seen in Fig 7.5.

In the former case the volume of dilution for the well was the main parameter. However, in this case the main parameter is the consumption of water for those nuclides, for which this pathway dominates the dose.



Relative contribution from dominant exposure pathways to total dose (D) and uncertainty (U) in the small well scenario (Case 1B).





Dominant parameters to the uncertainty in the small well scenario (Case 1 B).

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Case 2: lake scenario

In Fig 7.8 the percentual contribution to total dose and uncertainty for dominant pathways are shown.

The main pathways in this case are either through consumption of fish or agriculture products which are for simplicity lumped together in the figure as food. In this case Th-229 is also contradictory due to the main pathway via inhalation.

The main common parameters to the uncertainty are given in Fig 7.9. The expressions, sedimentation and migration in soil represent all parameters lumped together describing these processes respectively. For those nuclides with fish as dominating pathway the main parameters for the uncertainty are the amount of consumption, the uptake in fish and sedimentation. This latter process is decisive for the concentration in water, from which the uptake to fish is calculated. For Se-79 and I-129 with agricultural products as main pathway the main parameter is the migration in soil. For these nuclides many different pathways give the total dose illustrated as food. This means that additional contribution to the uncertainty arises from other parameters not illustrated in the figure.

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Dominant parameters to uncertainty in the lake scenario (Case 2) .

### Discussion

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In the previous sections the results of leakage of nuclides and decay chains to the biosphere, respectively, were shown. The uncertainty analysis performed showed the probable ranges of doses obtained from the turnover of nuclides in the actual ecosystem. One main parameter was the one describing the dilution of contaminated water within a well.

The individual doses from calculated leakages of the nuclides using a relatively high dilution volume of water are well below the annual doses man recieve from background radiation. If the available dilution volume decreases considerably the doses will increase. However, the extreme values used would have a very low probability to occur, because such a low volume can hardly support the necessary amount of water for a household including cattles.

Independent of future changes in climate, living habits etc and as long as there exists human beings, there is an obvious demand of fresh water. All the exposure scenarios studied are supposed to be likewise according to the present conditions.

One main difficulty is of course the long time periods for which leakage of nuclides may occur. During such periods the ecosystem will change. Within thousand of years lakes will dry up. The consequences for the exposure to man from an evolution of a lake was studied in another SKB-project. The results showed an increase of the doses for especially Pa-231 and Pu-239 with time (B Sundblad et al 1988).

In this study only fresh water recipients were considered. The transport of radionuclides with ground water make it possible to have different receptors depending on where the interface geosphere; biosphere is situated (eg. peat, soil and coast).

### Conclusion

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Individual doses from unit releases of long-lived nuclides and their decay chains leaking from a WP-cave repository for high level waste were calculated. Estimations of potential releases of activations and fission products showed that C-14, Se-79, Sn-126 and I-129 dominated the dose. For the actinides complete predictions using calculated releases to the biosphere were performed. However, only the leakage of the U-238 chain was calculated with solubility limitations which surely reduced the leakage rate to the biosphere considerable compared to the other chains. The Np-237 chain dominated the dose giving about 3.1.E-6 Sv/year, mostly caused by Np-237 and Th-229.

The dose-dominating activation and fission products will give maximum doses in the same order of magnitude as the actinides. However, the maximum doses from these nuclides will appear considerably earlier than for the actinides. For the latter nuclides maximum dose will appear after about one million year.

The highest doses for most of the nuclides are via consumption of water from the well. However, for those nuclides with a considerably concentration to biota the dominating pathway is via the lake water.

The uncertainty analysis showed that in the well, Case 1A, the main parameter is the volume of dilution in the well due to the wide range of the volume assumed. If the range is smaller as in Case 1B the consumption of water will be the main parameter to uncertainty. For those nuclides 1989-07-06

which have other dominant pathways the parameters describing these will dominate the uncertainty consequently.

The main parameters to the uncertainty in total dose for exposure via a lake are mainly those parameters describing the concentration in the water, in this study the sedimentation. In addition the fish pathway gives a considerable contribution to the uncertainty. The main parameters are the concentration factor and consumption of fish.

Further studies ought to be performed with special efforts on sensitivity analysis for other types of recipient areas as well as other important nuclides. Uncertainty analysis should be performed with variation in respective recipient areas. The purpose with further studies is to better understand the span in expected radiation doses as well as the uncertainties and processes important to the variation.

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# BIOPATH AND PRISM CODES BIOPATH-code

The mathematical method included in the BIOPATH code is based on compartment theory with firstorder kinetics. Therefore, the cycling and content of radioactive matter in different ecosystems are described by a system of first-order linear differential equations with constant or time varying transfer coefficients and a number of physically defined areas or volumes (which in this report are called reservoirs or compartments).

The premises are that:

- the outflow for reservoir "j" is solely dependent upon the quantity, y<sub>j</sub> of the radionuclide in that reservoir,
- the reservoir is instantaneously well mixed,
- all atoms, molecules or other elementary units have the same probability of leaving the reservoir

The amount of activity in a given reservoir is dependent on:

- 1) the outflow to and inflow from other reservoirs,
- 2) the source term for the reservoir, such as release to the reservoir or generation within it by decay.
- 3) radioactive decay

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This is expressed mathematically in vector form by:

$$\dot{Y}_{M}(t) = K_{M}Y_{M}(t) + Q_{M}(t) - \lambda_{M}Y_{M}(t)$$

for parent nuclides and

•

$$Y_{D_{n}}(t) = K_{D_{n}} Y_{D_{n}}(t) + \lambda_{D_{n}} D_{n^{-1}}(t) - \frac{\lambda_{D_{n}}}{D_{n}} D_{n^{-1}}(t) - \frac{\lambda_{D_{n}}}{D_{n^{-1}}} D_{n^{-1}}(t) + Q_{D_{n^{-1}}}(t)$$

for daughter nuclides.

The vectors Y and  $\dot{Y}$  refer to activity and activity changes per unit time in the different reservoirs of the system at time t. The coefficient matrices K (year-<sup>1</sup>) and Q(t) (activity year-<sup>1</sup>) describe the transfer rates between the reservoirs and sourceterm to the reservoir, respectively. The decay constant is

$$\lambda = \ln 2/t_{1/2}$$

where  $t_{1/2}$  is the physical half-life of the nuclide.

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### PRISM-code

The PRISM-system consists of three main parts. Firstly in PRISM 1 random model parameters are generated by using a systematic sampling method, Latin Hyper cube. As input to PRISM 1, the mean values, type of distributions, standard deviations and upper and lower limits must be given for each parameter. These data are used to define probability density functions. The Latin Hyper cube method, used to generate sets of parameter values from the given distributions, involves including all values within the ranges in the analysis. In addition correlations between model parameters can be taken into account no matter what distributions they are drawn from.

Secondly, in PRISM 2 model predictions, in this report dose calculations are made for each set of parameter values.

Finally PRISM 3 statistically evaluates and summarizes the joint set of model parameters and predictions.

The general statistics for the distribution of each parameter and the response of the model to this distribution contain the following:

- the arithmetic mean
- the standard deviation
- the coefficient of variation
- the geometric mean
- the percentiles (5, 25, 50, 75, and 95 %)
- the five highest values and the five lowest values, respectively

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Correlations between the model parameters and responses as well as between responses are also obtained from this last part of the analysis. Two correlations coefficients are calculated: firstly, the simple Pearson correlation coefficient and secondly Spearman R which is the correlation of the ranked values of the parameters and model responses. Associated with each correlation coefficient is percent COVAR. This represents the percent variance that one variable accounts for in another variable or response. In cases of correlated model parameters and responses, percent COVAR indicates the amount of variablility of the model response explained by the variablility of that model parameter.

The regression procedures are used to obtain the relationship between model parameters and model uncertainties.

The selection of parameters to be entered into the regression analysis is chosen among those which have the greatest improvement in the sum of squares of regression. Default values are given in the code but may be changed if chosen.

The output from PRISM 3 consists of all statistic information (intercept, slopes and mean values) required to write the regression equation for the relationships between model parameters and response. In these calculations the relative contribution to the total uncertainty from each parameter is also obtained. In this report the total dose received is obtained from summation via seven food pathways. This means that simple analytical equations can be used to characterize the relationships between total doses and each exposure pathway.

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#### RESERVOIR MASSES AND TRANSFER COEFFICIENTS

Transfer coefficients describing the transport of nuclides from water to sediment and the migration in the soil and groundwater are normally not possible to be found directly in literature. These transfer coefficients were estimated from physicochemical data on sorption, water and particulate transport etc in the same way as used in (Bergström et al 1983). A brief description of the methods and data used are given below. The masses of compartments and nuclide independent transfer coefficients are given in Table B.3 and B.5 respectively. The min and max values given are those used in the uncertainty analysis.

#### Water-sediment

For obtaining the transfer coefficient describing the transport of radionuclides from water to sediment, the following equation was used:

$$K = K \cdot S [h \cdot (1 + K \cdot SS)]$$
ws d m d y

where

K <sub>ws</sub>	<pre>= transfer coefficient water-sediment [y-1]</pre>
Kd	<pre>= distribution coefficient water, sediment =     concentration in solid/concentration in     liquid [m³/kg]</pre>
S	<pre>= sediment growth rate [kg/m<sup>2</sup>, y]</pre>
h <sub>m</sub>	= depth [m]
SS	= concentration of suspended matter in the water [kg/m <sup>3</sup> ]

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The following nuclide-independent data have been used:

	Best estimate	min	max
S	$= 1 \text{ kg/m}^2$ , y	0.1	3
h <sub>m</sub>	= 8 m	7	9
SS	$= 3E-3 kg/m^{3}$	1E-3	1E-2

The value of  $K_{d}$  is given in Table B.1 for each element.

# Migration in soils

The transfer coefficient describing the leakage from soil compartments was obtained from:

$$K_{s} = \frac{U_{w}}{R h_{i}} = \frac{U_{w}}{h_{i}} (1 + K_{d} \cdot S_{p} \frac{(1 - \varepsilon_{i})}{\varepsilon_{i}})$$

where

Ks	= transfer coefficient [y <sup>-1</sup> ]
U <sub>w</sub>	= water velocity through compartment [m/y]
h <sub>i</sub>	= depth of compartment i [m]
R	<pre>= retention = water velocity/nuclide   velocity</pre>
εi	= porosity of soil i [m <sup>3</sup> /m <sup>3</sup> ]
к <sub>а</sub>	= distribution coefficient [m <sup>3</sup> /kg]
s <sub>p</sub>	<pre>= density of soil particles [kg/m<sup>3</sup>]</pre>

The following nuclide-independent data were used:

	Best estimate	min	Max
U <sub>w</sub>	= 3 m/y	1	10
h <sub>1</sub>	= 0.3 m for the top soil	-	-
h <sub>2</sub>	= 1.7 m for the deep soil	-	-
sp	= 2.5.10 <sup>3</sup> kg/m <sup>3</sup>	1.5 E3	3.0 E3
ε_1	= $0.44$ for the top soil	0.2	0.6
<sup>ε</sup> 2	= 0.2 for the deep soil	0.1	0.6

The value of  $K_{d}$  is given in Table B.1 for each element.

# Groundwater to surface water

The transfer coefficient describing the leakage from the groundwater compartment was obtained from:

$$K_{g} = \frac{1}{RT} = \frac{1}{T} (1 + K_{d} \cdot S_{p} \frac{(1 - \varepsilon)}{\varepsilon})^{-1}$$

where

K g	=	transfer [y-1]	coefficient	groundwater	-	lake	

T = residence time for the groundwater in the actual compartment [y]

For other notations, see previous equation.

The following nuclide independent data were used:

Best estimateMinMaxT= 5 years110

The other parameters are set to the same as for deep soil.

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# Table B.1

Distribution coefficient  $[m^3/kg]$ , soil.

Nuclide	Best estimate	min	max	Ref
Ni	0.02	*	*	1
Se	0.01	0.001	1	1
Zr	30	*	*	2
Nb	10	*	*	7
Тс	0.005	*	*	2
Pd	0.02	*	*	4
Sn	0.1	0.05	0.5	4
I	0.3	0.1	1	2
Cs	1	*	*	1
Pb	0.1	*	*	4
Ra	0.5	*	*	5
Ac	1	*	*	4
Th	10	1	100	6
Pa	10	1	100	2
U	0.1	*	*	6
Np	0.1	0.01	1	2
Pu	50	*	*	3

\* Uncertainty analysis not performed

Ref	1)	Coughtrey	et	al,	1985
	2)	Bergström	et	al,	1983
	3)	Bergström	et	al,	1987
	4)	Bergström	et	al,	1985
	5)	Bergström	et	al,	1984
	6)	Grogan et	al,	. 198	36

7) Andersson et al 1979

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# Table B.2

Distribution coefficients [m<sup>3</sup>/kg], sediment- lake wate

Nuclide	Best estimate	min	max	ref
Ni	10	*	*	1
Se	5	1	10	1
Zr	5	*	*	1
Nb	100	*	*	1
Tc	0.1	*	*	1
Pd	10	*	*	2
Sn	50	10	100	1
I	0.3	0.1	1	1
Cs	10	*	*	1
Pb	0.05	*	*	2
Ra	10	*	*	3
Ac	10	*	*	2
Th	100	10	1000	4
Pa	100	10	1000	4
U	10	*	*	4
Np	10	1	100	1
Pu	100	*	*	1

\* Uncertainty analysis not performed

Ref

- 1) Coughtrey et al, 1985
- 2) Bergström et al, 1985
- 3) Bergström et al, 1984
- 4) Bergström et al, 1983

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# Table B.3

Transfer coefficient independent of element (year-1).

	Best estimate	Min	Max
Well - Local top soil (case A)	2.4E-4	5E-5	5E-4
Well - Local top soil (case B)	1.0E-2	5E-3	2E-2
Well - Lake (case A)	2.0	1	3
Well - Outflow (case B)	1	0.9	1.1
Lake - Regional top soil	4.4E-3	1E-3	1E-2
Lake - Outflow	3.0E-1	2E-1	4E-1
Top sediment - Lake	1.0E-3	1E-4	1E-2
Top sediment - Deep sediment	3.0E-2	1E-3	1E-1
Regional top soil - Regional atmosphere	1.0E-5		
Regional atmosphere - Global atmosphere	1.5E+2		

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# Table B.4

Element specific transfer coefficients (year-1) calculated with best estimate values.

Element	Top soil to Deep soil	Deep soil to well or Groundwater	Groundwater to lake	Lake water to sediment
Ni Se Zr Nb Tc Pd Sn I Cs Pu Np	1.6E-2 0.3 1.0E-4 3.1E-4 0.6 1.6E-2 3.1E-2 1.0E-2 3.0E-3 6.3E-5 3.1E-2 3.1E-2 3.1E-2	8.8E-3 1.7E-2 6.0E-6 1.8E-5 3.5E-2 8.8E-3 1.8E-3 6.0E-4 1.8E-4 3.5E-6 1.8E-3 1.8E-3 1.8E-3	1.0E-3 2.0E-3 7.7E-7 2.0E-6 4.0E-3 1.0E-3 2.0E-4 2.0E-2 2.0E-5 4.0E-7 2.0E-4	1.2 6.2E-1 6.2E-1 9.0 1.3E-2 1.2 5.4 3.8E-2 1.2 9.6 1.2
Th Pa Ra Ac Pb	3.1E-2 3.1E-4 3.1E-4 6.3E-3 3.1E-3 3.1E-2	1.8E-3 1.8E-5 1.8E-5 3.5E-4 1.8E-4 1.8E-3	2.0E-4 2.0E-6 2.0E-6 4.0E-5 2.0E-5 2.0E-4	1.2 9.6 9.6 1.2 1.2 6.2E-3

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# Table B.5

Reservoir masses (in dry weight or volume of water).

Local groundwater		
(Case A)	2.5 E5	m <sup>3</sup>
Local groundwater		
(Case B)	2.0 E3	m <sup>3</sup>
Local surface soil	1.7 E5	kg
Local deep soil	1.6 E6	kg
Lake water	3.2 E6	m³
Lake surface sediment	4.8 E7	kg
Regional surface soil	3.9 E7	kg
Regional deep soil	3.8 E8	kg
Regional groundwater	9.4 E4	m³
Regional atmosphere	3.7 E14	m³

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## UPTAKE IN FOOD CHAINS

The uptake of radioactive nuclides in different foods was calculated in the following way for the different pathways:

### Uptake in milk and meat

Radioactive elements in meat,  $U_k$ , and milk,  $U_m$ , originating from uptake via the following ecological paths of transport:

-	Root uptake to pasturage			
-	Deposition on pasturage from dry as well as wet deposition after irrigation			
-	Drinking water			
-	Intake of soil during grazing			

Thus,

 $U_{m}$  (in Bq per litre) =

=  $F_{m}((MC_{p} \cdot B_{p} + MC_{s}) \cdot C_{s} + MC_{1} \cdot C_{w} + MC_{p} \cdot (DEP \cdot MI \cdot C_{a} + K \cdot C_{w})),$ 

and

$$U_{k} \text{ (in Bq per kg)} =$$

$$= F_{k} ((MC_{p} \cdot B_{p} + MC_{s}) \cdot C_{s} + MC_{1} \cdot C_{w} + MC_{p} (DEP \cdot MI \cdot C_{a} + K \cdot C_{w})).$$

# Uptake in green vegetables

The concentration of radioactive elements in green vegetables,  $U_v$ , originates from two sources: the uptake of radioactivity via the root system, and deposition directly on the surfaces of the leaves.
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Thus,

 $U_{ij}$  (in Bq per kg) =

$$= B_{v} \cdot C_{s} + MI \cdot R \cdot (IRR \cdot C_{w} + DEP \cdot C_{a})$$

#### Uptake in grain and root vegetables

Uptake in grain,  $U_e$ , and root vegetables,  $U_r$  is assumed to take place primarily through the root system.

Thus,

 $U_{a}$  (in Bq per kg) =  $B_{q} \cdot C_{s}$ ,

and

 $U_r$  (in Bq per kg) =  $B_r \cdot C_s$ .

Uptake in eggs

Uptake in eggs, U<sub>e</sub>, comes from feeding the hens with contaminated grain and drinking water.

Thus,

 $U_{\rho}$  (in Ci per egg) =

$$= F_e(MC_g \cdot B_g \cdot C_s + MC_1 \cdot C_w).$$

#### Uptake in fish

Uptake in fish,  $U_{f}$ , is assumed to be directly proportional to the concentration in the water of the nuclides.

Thus,  $U_f$  (in Bq per kg) =  $B_f \cdot C_w$ .

Where:

U<sub>i</sub> = Uptake of one particular nuclide in foodstuff <u>i</u>. Given in Bq per unit of food (kg, litre or piece).

- 1 water consumption per animal
- k meat

,

- v green vegetables
- g grain
- r root vegetables
- f fish
- p pasturage
- s soil
- e egg
- B = Concentration factor of a certain nuclide for uptake via pathway n
- n = p soil → pasturage v soil → green vegetables g soil → grain r soil → root vegetables f water → fish
- F<sub>i</sub> = Distribution factor for a given nuclide for food-stuff <u>i</u>. Given in day per unit of food (kg or litre)
- C<sub>j</sub> = Concentration of a certain nuclide in reservoir j. Given in Bq per unit of reservoir
- j = w water

a air

s soil

MC<sub>i</sub> = Daily consumption of water, food-stuff (dry weight) and soil for cows in dominant transport links (l/day, kg/day). The consumption of water is 90 l/day, the STUDSVIK NUCLEAR

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consumption of pasturage is 16 kg dry weight/day ad the consumption of soil in connection with grazing is 0.3 kg/day. The mean consumption of water and grain for hen is 0.25 l/day and 0.11 kg/day, respectively.

- DEP = Deposition rate (m/day). DEP = 259 m/day for transfer from atmosphere to soil
- $MI_{i} = Mass interception factor (m<sup>2</sup> per kg)$  $MI_{p} = 1.8 m<sup>2</sup>/kg (dw) for pasturage$  $MI_{v} = 0.1 m<sup>2</sup>/kg for vegetables$
- IRR = Irrigation (1/m<sup>2</sup>, day)
  IRR = 0.4 for vegetables
- R = Average residence time on vegetation
  R = 22 days
- K = Lumped parameter considering retention of irrigated water during 6 occasions of irrigation

#### Table C.1

Concentration factors for transfer of nuclides from soil and water into food chains.

	Pasturage*	Cereals**	Green** vegetables	Root** vegetables	Fish***
Ni	9.5E-2	1.9E-2	1.9E-2	1.9E-2	100
Se	6.5	1.5	6.5	13	2000
Zr	8.5E-4	1.7E-4	1.7E-4	1.7E-4	60
Nb	4.7E-2	9.4E-3	9.4E-3	9.4E-3	10
Tc	1.0	9.0E-1	2.0E-1	1.0E-1	15
Pd	9.5E-2	1.9E-2	2.9E-2	1.9E-2	100
Sn	1.0E-1	3.6E-1	6.0E-2	4.6E-2	3000
I	1.0E-1	2.0E-1	2.0E-1	2.0E-1	50
Cs	1.0E-1	1.0E-2	2.0E-2	2.0E-2	10 000
Pu	9.0E-3	3.0E-5	1.0E-4	1.0E-4	30
Np	3.6E-2	4.6E-4	2.8E-3	2.0E-2	10
U	4.0E-3	1.0E-3	4.0E-4	8.0E-4	50
Th	5.0E-3	6.9E-4	5.0E-4	1.0E-3	30
Pa	3.0E-3	3.0E-3	3.0E-4	6.0E-4	10
Ra	2.0E-2	1.0E-2	7.0E-3	3.0E-3	25
Ac	1.0E-3	1.0E-4	1.0E-3	1.0E-3	25
Pb	4.5E-3	1.7E-2	1.8E-3	2.7E-3	100

- \* (Bq/kg dry weight)/(Bq/kg dry weight)
- \*\* (Bq/kg fresh weight)/(Bq/kg dry weight)
- \*\*\* (Bq/kg muscle fresh weight)/(Bg/l)

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# Table C.2

Variation of concentration factors for the uncertainty analyses. Triangular distributions were used. Best estimate values are given in table C.1.

	Se	Sn	I	Np	Th	Pa
Pasturage	1	1E-2	2E-2	7E-2	1E-3	1E-3
	70	1	0.3	0.2	1E-2	1E-2
Cereals	1	1E-2	5E-2	2E-4	1E-4	1E-3
	80	1	0.5	1E-2	1E-3	1E-2
Green vegetables	1	1E-2	5E-2	7E-4	1E-4	1E-4
	60	1	0.5	2E-2	1E-3	1E-3
Root vegetables	1	1E-2	5E-2	2E-4	5E-4	2E-4
	100	1	0.5	3E-2	5E-3	2E-3
Fish	1E+3	1E+3	10	1	1	5
	4E+3	6E+3	100	100	100	100

Min/Max

,

,

#### NOTES

#### Comments to table C.1

- Ni The data used are from the data base revision earlier performed (Bergström et al 1985). The values are mainly based on data from stable element. Data here are withdrawn from (Coughtrey et al 1985) and NUREG (NUREG-77) respectively.
- Se The values used were taken from the data base version (Bergström et al 1985).
- Zr The values used were taken from (NUREG-77) for concentration factors to plant and for fish from (Neuman 1985). These values are in the same range as those discussed in (Coughtrey et al 1985).
- Nb For the concentration factors to plant the values given by (NUREG-77) were used. However, lower values have been reported (Coughtrey et al 1985) but not considered due to bias of the values in order to not underestimate the exposure. The concentration factor to fish used is the value to muscle recommended by (Coughtrey et al 1985). Higher values of the factor are reported but this value refer to the whole body fish.
- Tc For the upptake to plant data were taken from (Bergström et al 1983). These data agree well with the values given by (IAEA 1987). The concentration factor to fish was taken from (Coughtrey et al 1983). A value of 30 is also given in the same reference but is probably for the whole fish.

- Pd Same values as for Ni was used due to the assumption of chemical similarity.
- Sn The values used are those reported in the revision (Bergström et al 1985).
- I The data for iodine are the same as previously used both for concentration to plant and fish (Bergström et al 1987). These values are within the range of well established data.
- Cs The concentration factors for plants are the same as used previously (Bergström et al 1987). New data are given by (IAEA-87) but the used data are within the ranges given in this draft. Because of the fact that the calculations are for an oligotrophic type of lake the concentration factor to fish is high. (Kolehmainen et al 1966, Evans 1988).
- Pu Plutonium was included in the data base revision earlier performed (Bergström et al 1985). These data for root uptake were compared with the values given in the IAEA draft (IAEA 1987), which has led to some minor changes. The bioaccumulation factor to fish was taken from (Agnedal 1978) and represent muscle. (Coughtrey et al 1985) gives a best estimate of 500 for fresh water fish, though this value obviously is for total fish.
- Np All root-uptake factors was taken from (Coughtrey et al 1985). For uptake to fish few data are available as default value 10 is often recommended in compilations (Bergström et al 1983) It is worth mentioning that it seems questionable that Np has a lower uptake factor than Pu.

- U The values used for root-uptake were taken from a literature survey, earlier performed (Bergström et al 1983). The value for uptake to fish is recommended as best estimate from (Poston et al 1986).
- Th For pasturage it is the geometric mean value estimated from values from (Ng 1981). For green vegetables and root vegetables the same value as for pasturage was used, corrected for fresh weight. For cereals the value used was the average value from Swedish investigations (Evans et al 1982). The value used for fish was taken from compilations (NUREG 77 and IAEA SS-57). However in a literature review performed by Poston et al a much higher value, 1000 is recommended. This value seems to be too high for a bone seeking nuclide, why it was not included.
- Pa All root-uptake factors were taken from (Bergström et al 1983). The best estimate for pasturage is obtained as the geometric mean for the values found. No values were found for other food-stuffs than pasturage, so values for cereals etc are the same as for pasturage, only corrected for fresh weight. Few data were unfortunately also available for uptake to fish. The value used was the one found in compilations such as in (IAEA SS-57).
- Ra Best estimate value for pasturage is the geometric mean of values found according to (Bergström et al 1983). Best estimate for cereals is mean value from Swedish investigations, (Evans et al 1982). The other

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rootuptake values are pasturage values corrected for fresh weight. The bioaccumulation factor to fish is the geometric mean value according to (Bergström et al 1983).

- Ac Due to the absence of data for actinium, data for americium was used instead, because of the chemical similarity between these nuclides. Best estimate values for root-uptake were estimated from (IAEA draft 1987). The bioaccumulation factor to fish was taken from (Bergström et al 1985). However Coughtrey 1985 suggests a factor of 200 for total body for all types of water. Americium will concentrate in bone, liver and kidney why a value of 25 seems reasonable for edible tissues of fish.
- Pb For terrestrial products all values were taken from, (Grogan 1985). For fish (Thompson et al 1972) gives a measured value of 100, which was used.

### Table C.3

Distribution factors for nuclides to milk and meat from total intake per day by the animal.

Element	พ่าษ	Moat
220m0110	/J	Meat
	day/1	day/kg
Ni	6.7E-3	2.0E-3
Se	3.0E-3	9.0E-4
Zr	5.0E-6	3.4E-2
Nb	2.5E-3	3.0E-7
Тс	1.0E-4	2.0E-3
Pd	6.7E-3	2.5E-3
Sn	1.2E-3	2.5E-3
I	1.3E-2	2.0E-3
Cs	8.0E-3	3.0E-2
Pu	1.0E-7	2.0E-6
Np	5.0E-6	3.0E-3
U	2.0E-4	1.0E-2
Th	5.0E-6	7.0E-4
Pa	5.0E-5	3.0E-3
Ra	3.0E-3	7.0E-4
Ac	2.5E-5	1.0E-2
Pb	2.6E-4	4.0E-4

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#### Table C.4

Variation of distribution factor used in the uncertainty analyses. Best estimate values given in Table C.3. Triangular distribution were used\*.

	Milk (day	y/l)	Meat (day	y/kg)
	Min	Max	Min	Max
Se	1E-3	1E-2	1E-4	1E-2
Sn	1E-4	1E-2	1E-4	1E-2
Np	1E-6	1E-4	2E-4	5E-3
Th	1E-7	1E-4	1E-4	1E-3
Pa	1E-6	1E-4	2E-6	5 <b>E-3</b>

\*

For I-129 log-normal distribution was used with standard deviation of 1.6 and 2.1 for milk and meat respectively. STUDSVIK NUCLEAR

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COMMENTS TO TABLE C.3

Element

Ni The factor for meat is given by (IAEA 1987), for milk no new value is given. The value used was taken from (Bergström et al 1987).

Se The factor for milk used was from (IAEA 1987). The value for meat is from (Bergström et al 1985).

- Zr The distribution factors used were taken from (Bergström et al 1983).
- Nb The value for milk was taken from (Bergström et al 1986). However for meat the value was taken from (IAEA 1987)
- Tc Earlier it was assumed due to lack of data that values for Tc uptake to milk were similar to I. Experiment carried out (Bondietti et al 1986) showed a much lower transfer to goat milk for Tc than for I. Because of that we used a lower factor, reflecting the relation between I and Tc. The transfer to meat was taken from (Bergström et al 1987).
- Pd Same values as for Ni were used due to the assumption of similar behaviour.
- Sn The values were taken from the revision of the biosphere data base (Bergström et al 1985). No new values were found in the literature.

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- I The data given by (IAEA 1987) were used. There are some minor differences from the values used previously.
- Cs The values given by (IAEA 1987) are almost the same as found in (Bergström et al 1985) why no changes of values were made.
- Pu Best estimate for milk was taken from the highest expected value according to (IAEA 1987). The best estimate for meat was also taken from the IAEA draft.
- Np Transfer to milk was estimated by (Ng et al 1981) in the absence of direct information. Values for the transfer to meat are reported to be within the interval 2E-4 - 5E-3. This implies a geometric mean value of 1E-3 which was used.
- U All values were taken from Bergström et al, 1983. These values are geometric mean values of ranges found in the literature.
- The transfer to milk is estimated in (IAEA SS-57) by Ng from human data in absence of relevant data. The distribution coefficient to meat is the geometric mean value of ranges found in the literature according to (Bergström et al 1983).
- Pa Best estimate for milk was taken from (Bergström et al, 1983) which refers back to (IAEA SS-57) where the value is estimated from (Ng et al 1981) in the absence of direct information.

Best estimate for meat is the geometric mean value of ranges found in the literature (Bergström et al, 1983).

Ra The values for milk and meat are geometric mean values of ranges found in the literature according to (Bergström et al, 1983).

- Ac In similarity with rootuptake factors the values are based upon information of americium (Bergström et al 1985). The values used are conservatively biased, especially for meat. Later information indicates values about a factor of 100 lower than the values used.
- Pb The behaviour of lead has not been studied to any great extent. However the values above are those recommended by Ng in (Nuclear Safety 23 1982).

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#### Table C.5

Consumption data for critical group and animals in the food chain.

	Best	Min	Max
	estimate		
Individuals			
Inhalation, m <sup>3</sup> /year	9438*	1000	1000
Drinking water, l/year	440	150	88
Milk, l/year	190	10	720
Meat, kg/year	55	25	100
Eggs, pc/year	200	-	-
Green vegetables, kg/year	25	10	100
Cereals, kg/year	75	25	200
Root-fruits, kg/year	75	25	200
Fish, kg/year	30	10	100
Cattle			
Drinking water, l/day	90.	75	110
Forage, kg/dag	16.	10	20
Soil, kg/day	0.3	0	1
Poultry**			
Drinking water, l/day	0.25	-	_
Cereals, kg/day	0.11	-	-

In the well scenario the time to cultivate the plot is limited and the inhalation during that time is assumed to be 96 m<sup>3</sup>/year.
 Not included in uncertainty analysis.

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# Table C.6

Variables in Case 1 B (small well) which differ from the values used in the base case.

Variable	Best estimate	Min	Max
Consumption of vegetable (kg/year)	10	5	25
Consumption of root vegetables (kg/year)	25	10	50
Equivalent consumption of water via irrigation of vegetables (litre)	1	0.5	5
Time for gardening the plot (h/year)	100	50.	200
Volume of the well (dilution) (liter)	2.0E6	1.5E6	2.5E
Time of turn over in the well (year-1)	1	0.9	1.1
Irrigation of the plot	1.0E-2	5E-3	2E-2

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Luis Moreno, Sue Arve, Ivars Neretnieks Royal Institute of Technology, Stockholm April 1989