

Long-term dynamics of a lake ecosystem and the implications for radiation exposure

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LONG-TERM DYNAMICS OF A LAKE ECOSYSTEM AND THE IMPLICATIONS FOR RADIATION EXPOSURE

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IMPLICATIONS FOR RADIATION EXPOSURE

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ABSTRACT

Long-term ageing and physical transformation of ecosystems may occur while a continuous leakage of radionuclides from a repository is going on. This will imply additional uncertainties as regards the consequences for exposure to man.

The turnover of nuclides during the ageing of a lake ecosystem and its successive development into agricultural land is simulated using a multicompartment system. Parameters of a major importance for the distribution and reconcentration of radionuclides supplied into the lake as surface inflow are identified. Seven radionuclides occurring in high-level waste are treated. These are I-129, Cs-135, Ra-226, Pa-231, U-234, Np-237 and Pu-239. The activity distribution is highly dependent on the sorption behaviour of the radionuclides. The major pools for radionuclide distribution are lake outflows 15 - 97 % (Pu-239 - I-129) and deep lake sediment 2 - 84 % (I-129 - Pu-239).

Performed dose calculations for different time periods of the lake evolution showed that the individual doses increase with a factor of hundred for Pu-239 during the life-time of the lake. For comparison doses have also been calculated for two different well scenarios in order to discuss the possibility of generic conversion factors from release to the biosphere and resulting individual doses. However, for all nuclides the obtained doses from exposure from a well situated in the discharge area to the lake were higher than for those obtained from the turnover of lake. For rough estimates the obtained doses can be used as standards when studying the impact on man from the turnover of long-lived radionuclides during the evolution of this type of ecosystem.

Keywords

lake evolution, radionuclide transfer, dose calculations, uncertainty analysis

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INTRODUCTION

1

The most hazardous radionuclides contained in high-level waste have such long physical half-lifes that they constitute a potential risk for man even in the distant future if they reach the biosphere in excessive amounts.

Leakage of nuclides from a repository may occur by the transport of groundwater and during such long time spans that the affected ecosystem may undergo changes with time. For example if a lake is the primary recipient for the nuclides this lake may be dried up during the time-span of potential leakages. An important question concerning the effects to man from such leakages is to determine the resulting changes in possible exposure which may occur.

The objective of this work is to assess the doses from a number of radicnuclides which may be continuously supplied from a high-level repository to a lake ecosystem during its successive ageing from lake to farming land.

This is made by applying the BIOPATH-code to a multicompartment model designed for a special ecosystem, which has been strictly investigated in earlier works leading to the determination of site specific parameter values (necessary for obtaining input data) to the model. The uncertainty in the results due to the uncertainty in the parameter values have been determined by using the PRISMsystem. Doses to critical groups have been calculated for two extreme stages of the lake.

Those doses are compared with those obtained from exposure from contaminated water reaching wells. Two cases have been studied where the first is representative for a well situated in the discharge area to the lake. The other

symbolizes a well created after the drying-up of the lake. This well is located in the soil, formerly sediment and the concentrations of the nuclides are taken from the concentrations in the pore water.

The evaluated nuclides are:

- I-129
- Cs-135
- Ra-226
- Pa-231
- U-234
- Np-237
- Pu-239

MODELLED ECOSYSTEM

2

Several lakes have been studied in the recipient evolution project. One of these is Lake Trobbofjärden, a eutrophic lake. This lake was a brackish bay at the Baltic coast 30 years ago. For further description, see /Sundblad 1986/.

Lake Trobbofjärden has been chosen for this model study, because it can be considered to be representative for most of the lakes in the southern part of Sweden.

The turnover of water and different elements in the lake, the sedimentation rate, the sediment properties from both chemical and physical point of view and the elemental composition in the sediment have been studied earlier within this project /Andersson 1987/, /Evans 1986/ and /Sundblad 1986/. Results from these reports have been used to identify important processes and to provide data for the simulation of the turnover of radionuclides during the recipient evolution.

3 DESCRIPTION OF THE CODES

Two codes have been used; one, BIOPATH, for solving the differential equations and calculate the doses to critical group and one, PRISM, for making uncertainty analyses due to the uncertainty in the input parameter values.

3.1 BIOPATH

BIOPATH is a general code based on compartment theory for solving the differential equations simulating the turnover of pollutants in the biosphere and the resulting effects to man. The exchange of nuclides or pollutants can either be expressed by a constant rate (per time unit) or be varying with time due to the available information. The same is applicable for the reservoir volumes or masses.

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

the outflow to and inflow from other reservoirs,
 any release within the reservoir, and
 radioactive decay

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

The vectors Y and \tilde{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⁻¹) and Q(t) (activity year⁻¹) describe the transfer rates between the reservoirs and release within 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.

In this work, when calculating the doses, the uptake to different food-stuffs is obtained from concentration factors applied for appropriate compartments. For further details of the code, see /Bergström et al, 1982/.

3.2 PRISM

PRISM is a system of programs designed to efficiently evaluate the uncertainty associated with model predictions as a result of uncertainties associated with model parameter values. The system is composed of 3 steps. Firstly, in PRISM1, the means, type of distribution, standard deviations and upper and lower limits of each parameter are used to define probability density functions.

Latin Hypercube sampling methods are used to generate the sets of random parameter values. Secondly, PRISM2 reads each set of parameter values, evaluates the model under consideration, and produces the model predictions. Finally, PRISM3 calculates all necessary statistic (intercept, slopes and mean values) to describe the regression between model parameters and responses.

In these calculations the relative contribution to the total uncertainty from parameters of importance is also obtained. Because the total dose consists of the sum of different exposure pathways, simple analytical equations have been used to characterize the relationship between total dose and each exposure pathway. For further information of the code, see /Gardner et al, 1983/.

4 DESCRIPTION OF THE MODEL

4.1 MODEL STRUCTURE

Based on the experience from previous studies, a model for the long-term transfer and distribution of radionuclides supplied to a lake ecosystem was evolved (Figure 4-1). It consists of two main components, the proper lake and the surrounding shore zone. Release of radioactive pollutants to the lake water is assumed to occur by inflow of contaminated water. The bottom sediment is divided into interstitial water and solid material. It is also divided vertically into two layers, separated by a redox-cline situated a few cm below the sediment surface. The gradual transition of the sediment in the shore zone to soil is also considered. The soil is divided into two compartments, consisting of material above and below the depth of the plough layer.



Figure 4-1 Model structure of the lake ecosystem.

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4.2 CONSIDERED PROCESSES

4.2.1 Sedimentation

The nuclides enter into the lake water, where they can be absorbed on particles. Most of these particles will settle. The settling velocity is usually described by Stokes law, see Appendix C. This law has been adopted for calculation of the settling velocity for particles of clay size.

The calculation of the transfer coefficient (kws) describing the transfer from water to sediment has been carried out according to Eq 4-1, where the content of suspended matter, distribution coefficient and depth of water are also included.

$$kws = \frac{u \cdot s \cdot Kd}{h}$$
 (Eq 4-1)

where

u	=	settling velocity (50 m/y)
S	=	suspended matter (0.01 kg/m ³)
Kd	=	distribution coefficient (m^3/kg)
h	=	depth (3 m)

This way of treating the flow of radionuclides from the water column to the bottom sediment constitutes a great simplification. The complexity of this transfer route is dependent on a lot of factors like particle size distribution, settling velocities, production rates and sizedependent scavenging properties of elements from the water column. This pathway also contributes substantially to the uncertainty of the calculated doses (see Chapter 6). However, because of lack of relevant data, we are at the moment forced to use Eq 4-1. Some of the problems concerning estimation of the loss of organic material from the water column are discussed in Appendix C.

4.2.2 Diffusion lake water - pore water

The concentration of soluble species is initially different in the lake and pore water of the sediment. However, this concentration gradient will decrease by diffusion. Lerman, 1978, describes this process as a part of a chemical model. The transfer coefficients (klp) describing this process are calculated according to Eq 4-2.

$$klp = \frac{\theta \cdot De}{h} \cdot \frac{1}{c} \cdot \frac{\Delta C}{\Delta z} \qquad (Eq \ 4-2)$$

where

θ	= water content (85 %)
De	= effective diffusion coefficient (m^2/y)
h	= water depth (3 m)
с	= nuclide concentration (Bq/m^3)
∆c/∆z	= concentration gradient $(Bq/m^3/m)$

The effective diffusion coefficient (De) is defined as

$$De = \frac{e \cdot Dw}{T^2}$$
 (Eq 4-3)

where

e = porosity
$$(0.85, m^3/m^3)$$

Dw = diffusion in water (m^2/y)
T = turtuosity $(1.2, m/m)$

The used Dw-values are given in Appendix A, Table A-4.

The tortuosity is defined as the average ratio of the actual "round-about" path to the apparent, or "straight" flow path. The tortuosity factor is sometimes defined as the inverse of the above /Hillel et al, 1971/.

The concentration gradients have been taken from the results presented by /Sundblad 1986/, where analogous data were accessible. The term

$$\frac{1}{c} \cdot \frac{\Delta c}{\Delta z}$$

is nuclide specific and varies between 1 to 20 for the handled nuclides.

4.2.3 Leakage pore water - lake water

The diffusion will affect the pore-water concentration as mentioned above. However, there are also other processes that govern the exchange between pore water and lake water as for example the compaction of sediment layer. This process has been described by the transfer coefficient (kpl) according to Eq 4-4.

$$kpl = \frac{\theta \cdot SR \cdot (s \cdot Kd + 1)}{h}$$
 (Eq 4-4)

where

θ	= water content in sediment (85 %)
SR	= sedimentation rate (0.003 m/y)
S	= suspended matter (0.01 kg/m^3)
Kd	= distribution coefficient (m ³ /kg)
h	= water depth (3 m)

4.2.4 Exchange between solid and water phase in sediment

The observed leakage of elements comparable to soluble nuclides from the sediments in Lake Trobbofjärden /Sundblad 1986/ has been used to calculate the transfer coefficient for non-sorbing species. The nuclide specific coefficients have been obtained by division with retention factors.

The retention factor is calculated according to $1+(1-\epsilon)*\circ p*Kd/\epsilon$. Notations see below (4.2.5) and Kd (Appendix D).

4.2.5 Transport in soil

Fresh soil is continuously built up in the contact zone between the solid ground and the shore zone. The resulting transfer (ks) within the soil from upper (30 cm) to lower compartment is described by Eq 4-5.

ks =
$$\frac{u}{d}$$
 · (1 + Kd · σp · (1- ϵ)/ ϵ)⁻¹
(Eq 4-5)

where

u	= water velocity (3.0 m/y)
d	= depth of soil (0.3 m)
ε	= porosity of soil $(0.2 \text{ m}^3/\text{m}^3)$
σp	<pre>= density of soil particles</pre>
	$(2.5 \cdot 10^3 \text{ kg/m}^3)$
Kd	= distribution coefficient (m ³ /kg)

The Kd-values describe the capability of adsorption for different nuclides. However, this adsorption depends on quite a lot of factors as for example type of soil, pH, Eh and ionic strength. Before chosing a Kd-value for the model calculation one thus has to consider the geochemical environment.

A literature review and some discussion of Kd-values have been made, presented in Appendix D.

4.3 TRANSFER COEFFICIENTS

The equations given above have partly been used to calculate the transfer coefficients for the total model system. However, there exists none or little information for the derivation or determination of some of the coefficients why experience from earlier calculations have been used to calculate these coefficients.

The transfer coefficients used for I-129, Cs-135, Ra-226, Pa-231, U-234, NP-237 and Pu-239 are given in Appendix A.

5 RESULTS: RADIONUCLIDE DISTRIBUTION

Calculations were performed for a period of 1 000 years to be able to follow how the activity released to a lake during its evolution was distributed within the ecosystem. The release was assumed to be 1 Bq/y into the surface water of the lake.

Three of the radionuclides, i.e. I-129, Cs-135 and Pu-239, were chosen for presentation of the activity distribution as a function of time. These radionuclides represent different sorption behaviour, see Table A-3 and cover the range of K_d -values for all nuclides.

5.1 WATER CONCENTRATION

The nuclide concentrations in lake water, shore zone and sediment are shown in Figures 5-1 to 5-3 for each nuclide, respectively. Steady-state condition is almost reached within 100 years for I-129 and Cs-135 in the lake water, while the concentration of Pu-239 increases slightly during the whole period.

The concentration in lake water varies within a factor of ten between the different nuclides. Because of the very high transfer to sediment of Pu-239 compared to Cs-235 and I-129 most of the released activity will be permanently fixed in the lake sediment. About 80 % of the activity of Pu-239 is found in the deep sediment (Table 5-1). The corresponding figures for I-129 and Cs-135 are 2 and 27 %, respectively.

The concentration of I-129 in lake water is around 6E-11 Bq/1 while the shore zone concentration is about three times higher (Figure 5-1). This is due both to the choice of source terms to the lake and to the shore zone

and on the slow exchange of water between these two compartments. The pore water concentrations reach higher levels than those of the lake water within 100 years.

The transfer of Pu-239 from water to sediment is much higher than the transfer for I-129. The results of this difference of transfer is clearly demonstrated in Figure 5-3 where the nuclide concentrations in the different water compartments are the opposite to the I-129 concentrations. see Figure 5-1. The lake water concentration for example is about 2E-11 Bq/1, while the shorezone level is about one order of magnitude lower.

All the other nuclides are between these two nuclides, see Figure 5-2 for Cs-135 as an example.

Table 5-1

Compartment	I-129	Cs-135	Ra-226	Pa-231	U-234	Np-237	Pu-239
Lake water	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1
Lake outflow	97.0	73.0	57.8	12.4	85.9	92.1	17.1
Lake sediment	<0.1	0.5	1.1	2.6	0.1	0.1	2.3
Lake sed deep	2.4	26.7	40.1	83.9	14.8	7.0	78.9
Soil upper	<0.1	0.1	0.1	0.1	<0.1	<0.1	0.1

Activity distribution (%) at 1 000 years.







Year

5.2 SOIL AND SEDIMENT CONCENTRATIONS

When a lake is ageing its volume is reduced /Agnedal et al 1984/. This reduction takes place both along the shore and in the deeper part of the lake. At the start of the exposure scenario, only a narrow strip of the shore is supposed to accumulate radionuclides. However, this shore strip increases according to a square function. After 1 000 years the total shore volume has increased to about 1.5 Mm³, which is equivalent to about 25 % of the total amount of sedimenting material. The remaining 75 % is built up in the deeper part of the lake.

The activity is transferred from the water to the sediment/soil compartment which is continuously growing. The nuclide flux to this compartment is almost constant during time, which means that the concentration will decrease because of the non linear growth of this compartment. This is also demonstrated in Figures 5-4 to 5-6, see soil (upper compartment).

The lake sediment concentration is highest for Cs-135 and Pu-239, while the shore zone sediment is highest for I-129. This is due to the high depletion of Pu-239 from the lake water to the sediment. This implies that a lower amount of this nuclide is brought to the shore zone compared with those nuclides which are more soluble in water.



Year



Year



Year

DOSE CALCULATIONS

6

In order to study when the maximal exposure to man will occur in the evolution of a lake, two different phases were chosen when making dose calculations with the codes. One phase (the lake) is defined as a young lake where no major changes due to drying-up has occurred, while the other phase (the soil) is the opposite when the lake has been dried up to such an extent that deeper layers of the sediment have been converted to farming land.

This phase has been chosen because it has been deemed to cause the highest accumulation of the activity in the ecosystem. However, there are some nuclides which can give higher concentration in the lake water with time, but this has been taken into account by varying the transfer rate from water to sediment to such an extent that they will cover the concentration in water obtained from the first phase of the modelling.

The exposure to man has been calculated by the BIOPATHcode in connection with the PRISM system to obtain the uncertainty in the results due to the uncertainty in the parameter values.

The doses for the time between these two phases, called "intermediate", have been estimated.

This intermediate phase represents the stage of the lake where the sediment in the shore zone has gradually been converted to soil.

6.1 PHASE "THE LAKE"

Since it is the concentration in the lake water which is of basic interest for this scenario, it was not deemed to be necessary to use the whole model earlier described for making the dose calculations, consequently a simplified system was used. The model consisted of only one reservoir for the water and two for the sediment. The soil was represented by two reservoirs; one for the upper plough-layer and one for the soil below this layer.

It was supposed that the water in the lake was used for irrigation of farming land in the viscinity of the lake during 100 years with 200 mm/year. It does not seem realistic to irrigate with surface water during a longer period without damaging the soil.

The used transfer coefficients are a summation of the transfer rates described earlier when it was appropriate, e.g. the resulting transfer from water to the sediment.

All the transfer coefficients were varied simultaneously within the code.

In Figure 6-1 the simplified system is shown and in Appendix B, Table B-1 the transfer rates with minimum and maximum values are summarized.



Figure 6-1 Compartment system for dose calculations.

6.2 PHASE "INTERMEDIATE"

The doses from this timeperiod have been estimated on the basis of the obtained concentrations from the BIOPATH-calculations.

There are also natural explanations why this phase could not be expected to cause any higher exposure than those calculated for the lake and soil case, respectively.

It is not possible that during the first hundred years the yield of this area could be enough for the annual consumption of food for critical groups.

6.3 PHASE "THE SOIL"

Based upon the calculations for the distribution of activity in the ecosystem the concentration of the nuclides in the deeper layers of the sediment at 1 000 years were chosen as source terms for the dose calculations. Uncertainty analyses were performed for the whole system for the nuclides Cs-135 and Pu-239. These calculations gave the requested mean value as well as the ranges corresponding to a 95 % confidence interval. The ranges for the other nuclides have been determined based upon estimates or the similarity with these nuclides. Those concentrations are shown in Table 6-1.

Concentration	I-129	Cs-135	Ra-226	Pa-231	U-234	Np-237	Pu-239
(Bq/kg)							
			<u></u>				
Best estimate	2.1E-9	7.3E-8	1.3E-7	3.7E-7	1.7E-8	1.7E-8	3.5E-7
Minimum	6E-10	3E-8	4E-8	1E-7	3E-9	5E-9	1E-7
Maximum	8E-9	2E-7	2E-7	6E - 7	6E-8	5E-8	6E-7

Table 6-1 Best estimate and ranges of the soil concentration used as source terms in the dose calculations.

Further it was assumed that after a period of 10 years this soil was used for agricultural purpose.

6.4 WELL SCENARIOS

In those scenarios the exposure to man is caused only by the amount of activity in the well-water.

The amount of water available for dilution is of course a very important factor for such a case. In these calculations this annual volume of water was varied within 1.E7 to 1.E8 1, mean value 5.4E7 1.

The assumptions for the calculated groundwater volume, affected by the withdrawal of water from the dug well (diameter 0.6 m), are the following:

The flow rate is 1 630 l/day (five persons utilize the well). A drawdown of 2 meters and a hydraulic conductivity of 5E-6 m/s will give an effective radius of about

240 meters (Davis, de Wiest, 1967). Thus the total affected groundwater volume, assuming an effective porosity of 10 %, will become 5.4E7 1.

For the other well scenario the basic concentrations in the water representing the well have been taken from the pore water concentrations obtained from the BIOPATH-calculations.

6.5 EXPOSURE PATHWAYS

Radioactive nuclides in the different reservoirs can reach man via different paths of exposure. Earlier calculations /Bergström U, 1983/ have shown that the dominant exposure paths for these nuclides are inhalation, consumption of food-stuffs and drinking of water. Internal exposure from food can take place via a number of links in the ecological transport chains such as

- uptake in crops via root uptake
- direct deposition which leads to temporary contamination of the surfaces of vegetation and/or translocation to the edible parts of the vegetation
- uptake via the food chain of grass-meat, grassmilk and grain-eggs
- cattle's consumption of water
- cattle's consumption of soil when grazing
- uptake in fish or other aquatic food-stuffs from surrounding water

In this scenario the exposure from the following exposure pathways has been considered:

- inhalation
- drinking water
- milk
- meat
- cereals
- vegetables
- root-vegetables
- fish

In Figures 6-2 and 6-3 there are schematic representations of how the different exposure pathways are calculated for the lake and soil case, respectively.

For the well scenarios the pathways considered are drinking-water for man and cattle and consumption of vegetables and root-vegetables grown on a small patch, irrigated with water from the well. They are illustrated in Figure 6-3.

The uptake for different food-chains is obtained from the expressions included in the BIOPATH-code. In Appendix B all the input parameter values with ranges or distributions are shown.



Figure 6-2 Exposure pathways.

Note:	ir	<pre>= irrigation</pre>
	r.u	= root uptake
	in	= ingestion



Figure 6-3


Figure 6.4

Exposure pathways for the critical group in the well scenario.

7 RESULTS OF DOSE CALCULATIONS

7.1 DOSE CALCULATIONS DURING THE EVOLUTION OF THE LAKE

Dominating exposure pathways and resulting doses originating from the contaminated lake ecosystem during its successive stages of ageing are presented for the seven radionuclides. The ranges of uncertainty in the dose estimations corresponding to a 95 % confidence interval, and the coefficient of determination (R2), which is the regression sum of squares divided by the total sum of squares and multiplied by 100 is also given for each scenario and nuclide. If the R²-value is high it is possible to draw conclusions such as where efforts would be concentrated to improve the model predictions. The results for each radionuclide are discussed below. The performed uncertainty analysis also gives information about which exposure pathways dominate the total uncertainty for the total dose.

7.1.1 Iodine-129

The change in total dose from I-129 with time is presented in Figure 7-1. At the start and endpoint, i.e. for the lake and soil case, the dose rate is of the same magnitude. The increase in the dose during the ageing of the lake is caused by a build-up in nuclide concentration within the shore zone. This may imply enhanced doses from milk and meat from cattle grazing close to the shore-line, and from drinking water collected at the edge of the lake.

In the <u>lake case</u>, the dominant exposure pathways were milk (36 %), fish (33 %) and vegetables (12 %) and it was also those pathways which dominated the uncertainty in the results with 64 %, 19 % and 7 % respectively.

In the <u>soil case</u> the contribution to the total dose was evenly distributed with about 30 % from each cereals, root vegetables and vegetables. Consequently those pathways gave also the dominant contributions to the uncertainty in the total dose, cereals dominated somewhat (34 %) followed by root vegetable (29 %) and vegetables (25 %).

The parameters dominating the uncertainty in the total dose are compiled below.

Table	7-1 The percentual contribut parameters to the uncert total dose for I-129.	ion of the ainty of f	e the
Case		(%)	R2
Lake	Turnover of water Consumption of milk	18 15	88
	Distribution factor, milk	13	
Soil	Concentration in soil	75	93
	Consumption of vegetables	3	
	Concentration factor, root vegetables	3	

Several parameters contributed with about the same order of magnitude to the uncertainty in the dose obtained for the lake case. The major contribution arouse from the parameter describing the turnover time of water in the lake, followed by the two parameters for describing the exposure from milk, namely the transfer to milk and the consumption of milk, respectively. The opposite was valid for the soil case where the concentration in soil totally dominated the uncertainty.



Lake intermediate bo

Figure 7-1

7.1.2 <u>Cesium-135</u>

For cesium there are no significant differences of the doses during the evolution of the lake (Figure 7-2). The results for the <u>lake case</u> are in good agreement with earlier calculations and experience, such as that the exposure from fish totally dominates both the exposure as well as the uncertainty with about 98 %. Exposure from milk and meat only contributes 2 % to the total dose as well as to the uncertainty.

In the <u>soil case</u> consumption of meat dominates (41 %) followed by milk (29 %). The expose from milk and meat gives about the same contribution (40 %) to the uncertainty.

The relative contribution from the parameters to the total uncertainty is shown in Table 7-2.

	dose for CS-135.		
Case		(१)	R2
Lake	Concentration factor, water-fish Consumption of fish Transfer water to sediment	67 17 9	95
Soil	Concentration factor, pasturage Distribution factor meat Concentration in soil	39 18 12	84

Table 7-2 The percentual contribution of the parameters to the uncertainty total dose for Cs-135.

For the <u>lake case</u> parameters describing the fish pathway dominate the uncertainty in agreement with the dominance for that exposure pathway to the total dose and uncertainty. There is much evidence how the uptake to fish is regulated by factors such as trophic level of fish and potassium content of the water. However, when making longterm prognoses like those for high level waste, it is not possible to have good knowledge about the characteristic of the future recipient.

The uptake of radioactive cesium in fish could also be even higher in nutrient-poor lakes, mostly situated in forested areas. The probability is, however, low that such areas will be converted to farming land.

In the <u>soil case</u>, the exposure is dominated by the animal products milk and meat in contrast to iodine which has the vegetable food as dominant exposure pathways. This is because Cs has a higher uptake to pasturage than for cereals and vegetables in combination with a high transfer to meat and milk.

The dominant source of uncertainty is due to the uncertainty in the root uptake factor for pasturage, reflecting the dominating exposure pathways milk and meat. The second and third most important parameters are the transfer of Cs to meat and the concentration in the soil, respectively. The calculations showed, in contrast to I-129, lower importance for the concentration in soil while the biological factors were of greater importance for the results for cesium, in the soil case.



Figure 7-2

7.1.3 <u>Radium-226</u>

The exposure from radium increases with about a factor of ten from the lake case to the soil case (see the results presented in Figure 7-3). It is not deemed possible that the phase between those so called extreme values could result in higher doses than those calculated, due to the great importance for the exposure from consumption of fish. In the <u>lake case</u> this pathway dominates both the dose and the uncertainty with 48 and 45 % respectively. Exposure through consumption of milk is the second one both as contribution to the total dose (26 %) and to the total uncertainty in the dose calculated (33 %). The third greatest contribution is from the exposure from consumption of water with about 17 % contribution to total dose as well as to the uncertainty.

In the <u>soil case</u>, exposure through consumption of cereals dominates both the exposure and the uncertainty (with 61 and 71 % respectively), followed by vegetables (20 and 22 %) and root-vegetables (14 and 6 %).

The relative contribution of parameters to the total uncertainty is shown below in Table 7-3.

	total dose for Ra-226.			
Case		(१)	R2	
Lake	Transfer water to sediment	73	94	
	Concentration factor, fish-water	10		
	Consumption of fish	3		
Soil	Root-uptake cereals	35	89	
	Concentration in soil	27		
	Consumption of cereals	14		

Table 7-3 The percentual contribution of the parameters to the uncertainty of the total dose for Ra-226.

The dominating parameter to the uncertainty in the lake case reflects the depletion of the activity in the water due to the transfer to the sediment. This transfer rate was varied to a large extent in the calculations which is consequently reflected in the results.

As can be seen from Table 7-3 the dominating contribution to the uncertainty caused by cereals emanates from the three parameters, describing the exposure from consumption of cereals, where the most important is from the rootuptake factor. The concentration in soil comes second. This concentration is caused by the transfer from water to sediment which implies that this transfer is of great importance for radium.





Figure 7-3

7.1.4 Protactinium-231

Results are presented in Figure 7-4. For Pa-231 the difference is more pronounced than for radium. The exposure increases with about a factor of twenty from the lake phase to the soil phase. Because the concentration in water remains fairly constant during time and the build-up of concentration in the shore zone is lower than in the proper lake, the doses will not increase until the transition of the sediment to soil.

In the <u>lake phase</u> it is the exposure pathways directly related to the activity in the water and not to the buildup in soil which dominates the exposure; the contribution from vegetables is due to the retention of irrigation water on the surfaces of the vegetation. The fish pathway dominates the exposure (63 %) followed by drinking water (27 %) and vegetables (9 %). Exposure from consumption of fish also gives the dominating contribution to the uncertainty (78 %). Thereafter, comes drinking-water (16 %) and vegetables (4 %).

In the <u>soil case</u> the exposure through cereals gives the dominant contribution to the total uncertainty (73 %) while inhalation only contributes (15 %) and root vegetables (6 %). The percentual contributions to total dose for these pathways are 54, 27 and 10 % respectively. The parameters contributing most to the uncertainty are given in Table 7-4 below.

In similarity with the other nuclides, with the exception of I-129, it is the transfer from water to sediment which dominates the uncertainty in the results. The bioaccumulation factor to fish is also important, followed by the dietary intake of fish.

Table	7-4 The percentual contribution o parameters to the uncertainty total dose of Pa-231.	f the of th	e
Case		(१)	R2
Lake	Water to sediment Concentration factor, fish-water	72 17	96
	Consumption of fish	4	
Soil	Concentration in soil	42	91
	Root-uptake factor, cereals	24	
	Consumption of cereals	18	

Pa-231 is considered to have low mobility with strong adsorption to particles. This is simulated by high transfer from water to sediment. However, because of the few data available we have varied this transfer rate with a factor 20. Because the transfer from water to sediment is of great importance for the concentration in water, it will totally dominate the uncertainty for Pa. The great importance of the transfer to the sediment is also reflected in the results for the soil case where the source term, namely the concentration in soil, dominates the uncertainty.

Thereafter in the lake case the concentration factor for fish follows while the fish amount consumed gives a much lower contribution to the uncertainty.

The contribution next in order to the uncertainty in the soil case is due to the root-uptake factor for cereals and finally the consumption of cereals. This reflects the higher uptake factor used for cereals than that for root-vegetables or vegetables. All the root-uptake factors for Pa had about the same ranges. As can be seen from Figure 7-4 the distribution of the resulting doses seems to belong to a normal distribution for the soil case.

To summarize, all the doses from Pa is very dependent upon the transfer from water to sediment and more knowledge about this process could decrease the uncertainty in the predicted results.





Figure 7-4

7.1.5 Uranium-234

The doses from uranium are, in similarity with cesium, not considerably affected by the scenarios. As can be seen from Figure 7-5 the ranges of uncertainty are about the same for the two cases. It is not likely that the exposure could be higher for the transition time than those calculated.

In the <u>lake case</u> the fish pathways dominates the dose (67 %) as well as the uncertainty (78 %). Thereafter drinking water appears with corresponding contributions of 21 and 16 % respectively. The other pathways give only minor contributions.

In the <u>soil case</u> the exposure through inhalation dominates the total dose in contrast to the results for the nuclides described earlier. Thereafter exposure from meat (21 %) and cereals (15 %) follow. It is the same pathways which dominate the uncertainty. The percentual contributions are 57, 24 and 14 % respectively.

The parameters contributing most to the uncertainty are given in Table 7-5 below.

	in the dose for U-234.		
Case		(१)	R2
Lake	Water to sediment Concentration factor, fish-water Consumption of fish	45 26 12	93
Soil	Concentration in soil Dust content in air Distribution factor, meat	73 13 3	92

Table 7-5 The percentual contribution of the parameters to the total uncertainty in the dose for U-234.

However, more parameters contribute to the uncertainty for uranium than for the nuclides mentioned earlier. Again the transfer from water to sediment is of importance and so is the bioaccumulation to fish.

There seems to be different information about the value of the bioaccumulation factor. According to /Agnedal, 1981/ a much lower value (5) than the one used for best estimate (50) is recommended. However, he also points out that data are scarce.

A lower value of this factor would decrease the doses for uranium independently of the lack of knowledge about the behaviour of uranium brought to a water recipient.

For the soil case it is, similar to Pa-231, the concentration in soil which dominates the uncertainty with as much as 73 %. The exposure through inhalation is calculated from the assumption that the dust particles in air come from dusting from soil associated with agricultural work. The doses obtained by inhalation is, therefore, to a big extent dependent on the concentration in soil. It is well recognized that it is difficult to find proper data for estimating this pathway. Naturally this pathway could be reduced considerably if modern agricultural technique is used, such as filters for the air entering the tractor cabin.

Briefly the same conclusions can be drawn for uranium as for the other nuclides studied, namely the big importance of estimating and making a correct description of the accumulation of uranium in the sediment.





Figure 7-5

7.1.6 Neptunium-237

There is also for Np-237 no considerable increase of the doses from the lake stage to the soil stage, see Figure 7-6. Because of the same reason as for uranium it has not been judged possible that the doses would neither increase nor decrease during the transition time.

As can be seen from the figure the ranges of uncertainties are well within a factor of ten. Consumption of fish dominates the exposure (67 %) in the <u>lake case</u> followed by consumption of water (24 %) and vegetables (8 %). For this case the fish pathway gives the dominating contribution to the uncertainty (83 %) followed by drinking water (13 %). Similarly to uranium the dominating exposure in the <u>soil case</u> is through inhalation (55 %). Consumption of vegetable food like root vegetables and vegetables are the next in importance with 17 and 13 % respectively.

These pathways percentual contributions to the uncertainty are 51, 27 and 10 % respectively

The dominating parameters to the total uncertainty are given in Table 7-6 below.

Table	7-6 The percentual contribution o parameters to the total uncer Np-237.	f the tainty	/ dose	for
Case		(%)	R2	-
Lake	Concentration factor, fish-water	37	93	
	Water to sediment	24		
	Consumption of fish	16		
Soil	Concentration in soil	48	80	
	Migration in soil	19		
	Amount of dust in air	9		

Similarly to cesium the bioaccumulation factor dominates the uncertainty, while the transfer from water to sediment is of minor importance for Np. Concerning the uptake to fish data are very scarce.

The assumption used here with a higher uptake factor than what is reported in compilations is also found in /Posten et al, 1986/.

They also suggest even higher values for fish species belonging to lower trophic values than herbivourous.

Concerning the transfer to the sediment Np is assumed to be quite soluble in the water because under aerobic conditions Np is found mostly as the stable NpO_2^+ ion. However, Np(V) can be reduced to much less soluble Np(IV) under anaerobic reducing conditions.

For the soil case the uncertainty in the results was mostly due to the uncertainty in the concentration in the soil, followed by migration in soil and amount of contaminated dust in the air. Due to the absence of pertinent information the concentration in the soil was varied within a factor of ten, see Table 6-1. This concentration is, as mentioned earlier, direct proportional to the transfer from water to sediment, which again reflects the big importance for describing this process realisticly with proper data. However, the natural variability in nature may well be within the ranges used. It is also interesting to note that Np-239 is the only nuclide for which the migration in soil gives a contribution to the total uncertainty. This is because Np-239 is relatively mobile in soil compared with the other actinides, which causes a reduction in the soil through the time span considered is only 10 years. This in combination with rather low

root-uptake factors and very low transfer for milk and meat explains why this process is of importance. The same discussions as held for uranium concerning the inhalation exposure pathway is also applicable for neptunium.

Better data are really needed in order to obtain better confidence in the results for Np-239 with regard to the fish and inhalation exposure pathways.



Figure 7-6

7.1.7 Plutonium-239

The results for Pu-239 showed the greatest increase when looking upon the results from the lake case compared to the soil case, see the results presented in Figure 7-7. It is not deemed reasonable that Pu can be assumed to cause any higher exposure than those calculated because of the great importance of the dose from inhalation, due to the same reasons as for U-234.

In the lake case the doses from ingestion dominate (fish 67 %, drinking-water 24 % and vegetables 7 %) while in the soil case both the contribution to the dose and to the total uncertainty is dominated by the exposure from inhalation with 96 % and 98 % contribution, respectively.

For the lake case the contribution to the uncertainty is more spread, i.e. fish (73 %), drinking water (24 %) and vegetables (7 %).

Likewise the results for the other nuclides the parameters contributing most to the uncertainty are given in Table 7-7 below.

ore /-/	paran the	neters to the uncertainty of the total dose for Pu-239.		
C	ase		(१)	R2
I	ake	Water to sediment Concentration factor, fish-water Consumption of fish	77 10 5	97
S	Soil	Dust content in air Concentration in soil Amount of air inhaled	55 34 3	94

Table 7-7 The percentual contribution of the

For the concentration in water causing the exposure in the lake case it is obviously the tranfer from water to the sediments causing the greatest source to the uncertainty.

For Pu an uncertainty analysis was performed for the total system (16 compartments). This analysis gave the ranges requested for the dose calculations. This range was within a factor of five, though the transfer from water to sediment was varied with a factor of 100. However, for Pu causing the greatest contribution to the total dose from inhalation, it was not the concentration in soil that dominated the uncertainty. It was instead the amount of contaminated particles in the soil that was the most important parameter.





Figure 7-7

7.2 RESULTS WELL SCENARIOS

The dominant pathway for all nuclides is from consumption of water, followed by vegetables. The results are presented in Table 7-8 for the well situated in the discharge area.

Table 7-8 Individual doses (Sv/y) and dominant exposure pathway for the well situated in the discharge area.

			Percentual to the dos	l contribution se
Nuclide	Mean	Ranges	Drinking water	Vegetables
I-129	4.2E-12	1.1E-12 - 1.6E-11	29	31
Cs-135	9.5E-14	2.5E-14 - 3.9E-13	25	39
Ra-226	7.8E-12	2.3E-12 - 2.8E-11	53	30
Pa-231	3.8E-10	1.0E-10 - 1.4E-9	74	24
U-234	5.8E-12	1.6E-12 - 7.9E-11	65	21
Np-237	2.1E-1]	5.7E-12 - 7.9E-11	72	2.4
Pu-239	2.1E-11	5.7E-12 - 7.6E-11	73	24

The value for dilution is the most important factor concerning the accuracy in the predictions due to the uncertainty in the parameter values. For all nuclides this contribution is about 75 %. Thereafter other parameters like consumed amount of water and vegetables, and retention of the surfaces of vegetation give only minor contributions.

Exactly the same parameters will of course be of importance for a well situated in the former sediments. However, the doses obtained are lower becuase of a lower concentration in this water than for the first well.

8 COMPARISON OF DOSES

In Table 8-1 the doses are presented in time-order of the different scenarios studied.

Table 8-1

Nuclide	Well*	Lake		Soil	Well**
I-129	4.2E-12	2.4E-14		1.7E-14	2.1E-13
Cs-135	9.5E-14	5.3E-15		1.9E-15	1.5E-14
Ra-226	7.8E-12	1.8E-14		1.5E-13	2.4E-13
Pa-231	3.8E-10	1.0E-13	>	8.0E-12	2.5E-12
U-234	5.8E-12	3.1E-14		1.8E-14	1.8E-13
Np-237	2.1E-11	1.5E-13		3.6E-13	6.4E-13
- Pu-239	2.0E-11	1.0E-14	<u> </u>	7.9E-13	2.5E-14

Situated in the discharge area to the lake.
Situated in the soil former sediments.

As can be seen from the table the doses from the "first" well is in general two orders of magnitude higher than for the lake or soil case.

For the "second" well case there is no general relation between the scenarios. For more soluble nuclides the exposure from this well is higher than those for the different stages of the lake.

DISCUSSIONS

9

The calculated doses show only minor variations during the ageing of the lake. However, for nuclides with low mobility such as Pa-231 and Pu-239 there is a substantial increase at the final stage of the lake development due to the fixation in sediments. These sediments which due to drying-up became available for agricultural purposes at the end stage of the lake.

For most of the nuclides the transfer from water to sediment is of major importance for the final results. In addition, the uptake to fish is identified as an important process, especially as data often are scarce and even contradictory. Better data are also required for making more realistic calculations of the exposure through inhalation, especially for Pu. For the studied scenario the performed uncertainty analysis showed that the uncertainty in the results was mostly due to the uncertainty of simulating the turnover of the nuclides in the ecosystem.

For all nuclides the doses increased with about a factor of hundred for a well situated in the discharge area to the lake. No consideration has been taken for any reduction due to filtration of the water or adsorption to particles. It is assumed that the nuclides are in a soluble form in the water. Concerning wells another important question is the probability of the localisation of the well.

In general, the ranges of uncertainty are within a factor of ten, though the parameter values are varied within a factor of 10 to 100.

The calculations for a eutrophic lake ecosystem showed that the potential maximum exposure is not specifically sensitive to the stage of evolution of the lake. A set of conversion factors between release and dose ("standards")

can be set up for that type of lake ecosystems. These "standards" will also include uncertainties.

There are other types of ecosystems. However, it is not possible, from the present calculations, to formulate corresponding "standards" for other ecosystems. The ecosystems of interest are:

- Oligotrophic/dystrophic lake
- Peatland
- Discharge area, lake sediments
- Discharge area, agricultural land

From the achieved knowledge of dose calculations from the eutrophic lake ecosystem it is possible, by complementary information, to calculate the corresponding "standards" for the above mentioned ecosystems.

In the model calculations simplifications have been made that can affect the results. For example, correlations between parameters and the build-up of daughter nuclides have not been considered.

Finally, it would also be of value to compare the results from this approach for modelling the long-term dynamics of a lake ecosystem and the implications for the doses with the results from the more static model earlier used.

To ensure the confidence in model results they need to be tested against independant data sets. Unfortunately it is impossible to find such data for testing the whole of the model described above. However there may be possibilities foe evaluation of subparts of it like transfer to sediment or maybe most possible the uptake to fish. In the absence of "validation" it is important to make intercomparisons of different models addressing the same problem. In the BIOMOVS study an international effort for testing and intercomparison of radioecological models is going on. One of the scenarios deals with modelling the tunover of Ra-226 and Th-230 during the evolution of the lake Trobbofjärden. The model described above has participated in this intercomparison. In another BIOMOVS scenario participants were asked to model the turnover of these above mentioned radionuclides in a hypothetical lake. Concentrations in water, sediment and edible tissues of fish were requested. One of the main conclusions from this latter scenario was the obvious demand to find independant data sets for testing the uptake to fish of Ra-226 and Th-230. The complete dose calculations carried out in this study confirm this because of the big importance of the fish exposure pathway.

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Appendix A.1

APPENDIX A

PARAMETER VALUES FOR CALCULATING THE DISTRIBUTION OF THE NUCLIDES

Number	Туре	Mass
		(kg)
1	Lake water	9.30E+09 decreasing
2	Shore zone water	1.16E+08
3	Shore zone water sediment minerogenic	2.64E+06
4	Soil upper (0 - 30 cm)	1.51E+03 increasing
5	Lake water outflow	1.00E+03
6	Lake sediment pore water upper (0 – 2.5 cm)	7.00E+07
7	Lake sediment pore water lower (2.5 - 10 cm)	2.00E+08
8	Soil lower	1.40E+03
9	Shore zone sediment deep (> 10 cm)	5.64E+07
10	Lake sediment solid upper (0 - 2.5cm)	6.60E+06
11	Lake sediment solid minerogenic lower	6.50E+07
12	Lake sediment solid organogenic upper	1.20E+06
13	Shore zone sediment pore water	1.78E+06
14	Shore zone sediment organogenic	1.10E+06
15	Lake sediment deep (sink)	1.00E+03
16	Shore zone sediment deep (sink)	1.20E+03

Table A-1 Compartment description.

NP224 EA
Compan	rtments	Nuclide						
From	То	I-129	Cs-135	Ra-226	Pa-231	U-234	Np-237	Pu-239
1	2	1.6E-03						
1	5	1.3E+00						
1	6	4.5E-02	1.8E-01	4.2E-02	3.0E-02	3.0E-02	3.0E-02	4.5E-03
1	10	1.7E-02	8.4E-01	1.7E+00	1.7E+01	1.7E-01	1.7E-01	1.3E+01
]	12	2.6E-03	1.3E-01	2.6E-01	2.5E+00	2.5E-02	2.5E-02	1.9E+00
2	1	1.3E-01						
2	3	5.0E-02	2.5E+00	5.1E+00	5.0E+01	5.0E-01	5.0E-1	3.8E+01
2	13	1.4E-01	6.0E-01	1.3E-01	1.0E-01	1.0E-01	1.0E-01	1.5E-02
2	14	7.7E-03	3.8E-01	7.8E-01	7.5E+00	7.5E-02	7.5E-02	5.7E-01
3	4	8.0E-03						
3	9	3.3E-02						
3	13	1.0E-05	2.0E-07	2.0E-07	2.0E-08	1.0E-07	2.0E-06	2.0E-09
4	8	1.0E-02	2.0E-03	2.0E-03	2.0E-04	1.0E-02	2.0E-02	2.0E-05
6	1	1.0E-03						
6	7	3.0E-02						
6	10	1.0E-03						
7	6	1.0E-03						
7	11	1.0E-03						
7	15	3.3E-02						
9	16	1.0E-02						
10	6	1.0E-04	2.0E-07	2.0E-07	2.0E-08	1.0E-07	2.0E-06	2.0E-09
10	11	1.0E-01						
11	7	1.0E-05	2.0E-07	1.0E-07	2.0E-08	1.0E-07	2.0E-06	2.0E-09
11	15	3.3E-02						
12	10	5.0E+00	5.0E+00	5.0E+00	5.0E+00	5.0E+00	5.0E+00	5-0E+00
13	2.	3.0E-03						
13	3	1.0E-03						
13	9	1.0E-01						
14	3	5.0E+00						

Table A-2 Transfer coefficients for I-129, Cs-135, Ra-226, Pa-231, U-234, Np-237 and Pu-239.

Table A-3 Distribution coefficients (m³/kg).

Nuclide	Sediment	Soil	
I-129	0.1	0.01	
Cs-135	5.0	0.50	
Ra-226	10.0	0.50	
Pa-231	100.0	5.00	
U-234	1.0	0.10	
Np-237	1.0	0.05	
Pu-239	75.0	50.0	

References

I- and Cs-values from	Andersson 1987
Ra values from	Bergström, Andersson, Röjder 1983
Pa values from	Estimations at Studsvik
U values from	Andersson 1983
Np- and Pu-values from	Coughtrey 1985

Table A-4 Diffusion coefficient (m^2/y) .

Nuclide	Dw
I-129	3.2E-2
Cs-135	3.2E-2
Ra-226	2.5E-2
Pa-231	1.6E-2
U-234	1.6E-2
Np-237	1.6E-2
Pu-239	1.6E-2

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Dw-values from Andersson 1983

Appendix A.5

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APPENDIX B

INPUT PARAMETER VALUES FOR THE DOSE CALCULATIONS

In this Appendix the input parameter values used for the dose calculations are shown.

Most of the parameters are either assumed to belong to triangular or logtriangular distributions.

References to the values used and/or comments to the choice of values are given as comments after the Tables.

Transfer coefficient	I	Cs	Ra	Pa	U	Np	Pu
Water - sediment	6.5E-2	9.7E-1	2.0	2.0E1	4.6E-1	2.0E-1	1.3E1
min	6.5E-3	9.7E-2	2.0E-1	2.0	4.6E-2	2.0E-2	1.3
max	6.5E-1	9.7	2.0E1	2.0E2	4.6	2.0	1.3E2
Water - soil	2.0E-2						
min	1.0E-2						
max	3.0E-2						
Outflow	1.3	1.3	1.3	1.3	1.3	1.3	1.3
min	7.5E-1						
max	2	2	2	2	2	2	2
Sediment - water	1.0E-3						
min	5.0E-4						
max	5.0E-3						
Upper sediment - deeper sediment	1.3E-1						
min	4.0E-2						
max	4.0E-1						
Deeper sediment - upper sediment	1.0E-3						
min	5.0E-4						
max	5.0E-3						
Deeper sediment - sediment sink	3.3E-2						
min	1.0E-2						
max	1.0E-1						
Upper soil - deeper soil	1.0E-2	2.0E-3	2.0E-3	2.0E-4	4.0E-3	2.0E-2	2.0E-5
min	1.0E-3	5.0E-4	2.0E-4	2.0E-5	1.0E-4	2.0E-3	1.0E-6
max	5.0E-2	1.0E-2	2.0E-2	2.0E-3	1.0E-2	2.0E-1	5.0E-4

Table B-1 Transfer coefficients for dose calculations (unit y^{-1}), best estimate and ranges.

Element	Pasturage*	Ranges	Cereals	Ranges	Leafy vege- tables	Ranges	Root vege- tables	Ranges
- <u> </u>								
I	2.0E-1	2E-2 - 3E-1	2.0E-1	5E-2 - 5E-1	2.0E-1	5E-2 - 5E4	2.0E-1	5E-2 - 5E-1
Cs**	1.0E-1		1.0E-2		2.0E-2		2.0E-2	
Ra	1.0E-1	1E-2 - 6E-1	1.0E-2	1E-3 - 5E-2	1.2E-2	1E-4 - 4E-2	3.0E-3	1E-3 - 1E-2
Pa	3.0E-3	1E-3 - 1E-2	3.0E-3	1E-3 - 1E-2	3.0E-4	1E-4 - 1E-3	6.0E-4	2E-4 - 2E-3
U	4.0E-3	1E-3 - 1E-2	1.0E-3	1E-4 - 1E-2	4.0E-4	1E-4 - 1E-3	8.0E-4	2E-4 - 2E-3
Np	3.0E-2	7E-3 - 2E-1	3.0E-3	2E-4 - 1E-2	3.0E-3	7E-4 - 2E-2	6.0E-3	2E-4 - 3E-2
Pu	9.0E-3	1E-3 - 2E-2	1.0E-4	4E-5 - 3E-4	1.0E-4	1E-5 - 1E-3	3.0E-4	1E-4 - 1E-3

Table B-2 Root uptake factors for several types of nutrients (Bq/kg of nutrient f.w. per Bq/kg of dry soil).

* For pasturage the values are given in dry weight.

* Lognormally distributed with standard deviation 2.48, 1.8, 1.9 and 1.9 for the nutrients, respectively.

Element	Distribution factor milk	Ranges or standard deviation	Distribution factor meat	Ranges or standard deviation
	(uay/1)		(uay/kg)	
1*	1.0E-2	1.6	2.0E-3	2.1
Cs*	7.0E-3	1.6	3.0E-2	2.1
Ra	3.8E-3	7.1E-4 - 8.4E-3	7.0E-4	5.0E-4 - 1.0E-3
Pa	5.0E-6	1.0E-6 - 1.0E-4	3.0E-3	2.0E-6 - 5.0E-3
U	2.0E-4	4.0E-5 - 6.0E-4	1.0E-2	5.0E-3 - 3.0E-2
Np	1.0E-5	5.0E-6 - 1.0E-4	1.0E-3	2.0E-4 - 5.0E-3
Pu	1.0E-7	5.0E-8 - 1.0E-6	2.0E-6	1.0E-6 - 5.0E-6

Table B-3 Distribution factors for milk and meat.

* Assumed to be lognormally distributed.

Element	Best estimate	Ranges
I	50	10 - 100
Cs	1.300	200 - 5.000
Ra	25	10 - 100
Pa	10	5 - 100
U	50	5 - 100
Np	30	10 - 100
Pu	30	10 - 100

Table B-4 Concentration rates to fish (Bq/kg per Bq/1).

Table B-5 Consumption data, triangularly distributed.

	Best estimate	Ranges
Water (1/year)	440	150 - 880
Milk (l/year)	190	50 - 720
Meat (kg/year)	55	20 - 100
Cereals (kg/year)	75	25 - 200
Leafy vegetables (kg/year)	75	25 - 200
Root fruit (kg/year)	75	25 - 200
Fish (kg/year)	30	10 - 50

Table B-6 Inhalation data.

	Best estimate	Ranges
Amount of particles in the air (kg/m^3)	5.0E-5	1E-5 - 1E-4
Dave of exposure (dave)	30	8 - 12
bays of exposure (days)	JU	<u>ر</u> ر - ر_

ł

		Weighted do: (Sv/i	se equivalents Bq)
Nuclide	Half- ³ lif	e(y)Ingestion	Inhalation
1-129	1.6E7	9.8E-8	4.7E-8
Cs-135	2.3E6	1.9E-9	1.2E-9
Ra-226	1.6E3	3.3E-7	2.1E-6
Pa-231	3.3E4	2.2E-5	3.4E-4
U-234	2.4E5	3.0E-7	3.6E-5
Np-237	2.1E6	1.2E-6	1.3E-4
Pu-239	2.4E4	1.2E-6	1.4E-4

Table B-7 Dose conversion factors. Sum of weighted commited dose equivalents.

References

ICRP30, ICRP40, Johansson, L, 1982.

COMMENTS TO TABLE B-2

Element

- I Best estimate values are taken from Bergström et al, 1985. The ranges for pasturage are taken from Bergström et al, 1983. Due to lack of information about values for the other nutrients they are varied with a factor of five.
- Cs All values are taken from Bergström et al, 1983.
- Ra Values for pasturage are taken from Watson et al, 1984. Best estimate for cereals is taken from Evans et al, 1982. However, the ranges applied are higher than those described in Evans et al, 1982. For vegetables all values are taken from Watson et al, 1984. The best estimate for rootvegetables is taken from Bergström et al, 1983 and the ranges are estimated to be within a factor of ten.
- Pa All best-estimate values are from Bergström et al, 1983. Data are very sparse while the ranges used are set to about a factor of three lower and higher, respectively.
- U All best-estimate values are from Bergström et al, 1983. The ranges used are estimated to be within a factor of ten. However, this might be an overestimation of the ranges for cereals, according to Evans et al, 1982. They give a standard deviation of 1 for this factor.
- Np For pasturage best estimate and ranges are taken from Bergström et al, 1983. Best estimate for vegetables and root vegetables are also taken from that reference and due to the abscense of data the ranges are put according to the values for pasturage. The low value in this reference for cereals is not used, because there are clear evidence that this value would underestimate the uptake to cereals. Because of that the same best estimate as for vegetables has been used, with quite large ranges.
- Pu For pasturage best estimate is taken from Bergström et al, 1987. The ranges used for pasturage cover those given in IAEA draft for grass and fodder. For cereals the best estimate is also taken from Bergström et al, 1987 and the ranges are taken from the IAEA draft. For vegetables the best estimate is from Bergström et al, 1987. The ranges used are somewhat higher than

those given in the IAEA draft. For root vegetables (also including potatoes) the best- estimate value corresponds well with the most expected value for different conditions in the IAEA draft.

The ranges are set so they will not be an underestimation of the ranges found in the literature. COMMENTS TO TABLE B-3

Element

- I Best estimate is taken from Hoffman F O et al, 1979. The standard deviation is also about the same range as reported in that reference.
- Cs The parameters are supposed to be lognormally distributed and all values are taken from Bergström et al, 1983.
- Ra The values for milk are all taken directly from Watson et al, 1984, so is also the best-estimate value for meat while the ranges for meat are taken from Bergström et al, 1983.
- Pa Best estimate for milk is taken from Bergström et al, 1983 which refers back to IAEA Series 57 where the value is estimated from Ng in the absence of direct information. This has implied that the ranges used are based on estimates.

Best estimate for meat is also taken from Bergström et al, 1983. The ranges used included also a single value based on a similar behaviour of Pa as for Am and Cm.

- U All values are taken from Bergström et al, 1983.
- Np Coughtrey et al, 1985 reports values for milk to be in the range of 5E-6 to 1E-4. This range has been used with an estimation of the best value. The values for meat are taken from Bergström et al, 1983.
- Pu Best estimate for milk is taken from the highest expected value according to IAEA draft. The ranges used include higher values than those reported in the report mentioned above. However, the ranges are of no interest, because of the low transfer to milk.

The best estimate for meat is also taken from IAEA draft, which also gives ranges which are higher than those used in this report. However, this does not affect the results because the exposure through meat is of no importance for Pu.

COMMENTS TO TABLE B-4

Element

Ι

In earlier work, like Thompson et al, 1981, a value of 15 was recommended for fresh-water fish. Also Neumann, G, 1985 recommends a low value of 15. Coughtrey et al, 1983 recommended a value of 50, which has been used as best estimate in this study. However, according to Poston, T M, et al, 1986 the uptake would be substantially higher, 200 for muscle of piscivorous fish and 500 to omnivorous and planktivorous species. Because those values were not found when the calculations were made, they have not been taken into account. But is is worthwile remembering this when discussing the results for iodine.

The ranges used are estimated, but the lower range corresponds well to the values given from Thompson et al, 1972.

Cs According to Vanderploeg et al, 1975 the uptake to non-piscivorous fish can be written as 5×10^{-7} (K) for clean water, where K = stable potassium concentration of water in ppm.

> The concentration of potassium in Lake Trobbofjärden is about 4 ppm from Andersson, K, 1987. This would give a factor of about 1250. The same value is recommended in Bergström et al, 1983 as best estimate. The ranges used are taken from the latter reference, remembering that the lake in the study is of eutrophical character.

- Ra The values used are taken from Bergström et al, 1984.
- Pa Best-estimate value is taken from Bergström et al, 1983. The ranges set are estimated based upon the fact that it is not probable that the uptake of Pa is higher than that for Pu.
- U Earlier reported values for uptake to fish are low, in the range of 2 - 10 according to the literautere review made in Bergström et al, 1983. However, Poston, T M et al, 1986, reccomend a value of 50. This has been used as best estimate with ranges covering the values reported earlier.

Np Very few data are available for the uptake to fish. In compilations 10 is often recommended as default value.

In order not to underestimate this pathway the same values as for Pu have been used. It seems also reasonable that the uptake of Np should not be considerably lower than that for Pu.

Pu The values used correspond to the values given in Agnedal, P-O, 1978. The ranges used are somewhat less scattered than those given in the reference.

APPENDIX C

FACTORS AFFECTING PHYTOPLANKTON LOSS FROM THE WATER COLUMN

The most essential requirement of phytoplankton is some means of prolonging suspension in the upper, illuminated layers of the water column. A low overall sinking rate minimizes the opportunity to escape from the turbulent-mixed layer into lower, non-turbulent layers. The various groups represented in marine and freshwater phytoplankton achieve a variety of adaptive mechanisms for depressing the sinking rate. Those selected by most planktonic algae favour either small size, reduced excess density or devices for increasing frictional resistance with the surrounding water. In addition, many planktonic organisms retain a behavioural or physiological control over their sinking behaviour.

Effects of size

Microscopic size is perhaps the most widespread adaption in order to achieve a low sinking rate. The calculated radii are in the range $1 - 285 \mu$ for a number of planktonic algae /Reynolds, 1984/. Besides, the effective size of many planktonic algae is also increased by colony formation. The effect of size on velocity has been demonstrated by Smayda /1970/ for several marine species of centric diatoms. The maximum velocity reported for any uncellular algae is 6 mm s⁻¹, observed for empty frustules of the large marine diatom <u>Ethmodiscus rex</u>, having a diameter of approximately 1 mm /Smayda, 1970/. However, the settling velocities of most other diatoms considered by Smayda /1970/ were 1 - 2 orders of magnitude less than that of <u>E</u>. <u>rex</u>.

Settling velocities

The settling properties of planktonic cells and colonies are determined by the same forces which govern the movements of inert bodies in viscous fluids. The terminal velocity of a spherical body 1s given by the Stokes equation

$$v_{g} = \frac{2 gr^{2} (\rho' - \rho)}{9 r_{l}}$$
 (Eq 1)

g	= the gravitational acceleration (m s^{-2})
η	<pre>= the coefficient of viscocity of the fluid medium (kg m⁻¹s⁻¹)</pre>
ρ	= the density of the medium (kg m^{-3})
ρ'	= the density of the particle (kg m^{-3})
r	= the radius of the particle (m)

The term $(\rho'-\rho)$ is known as the excess density; when it is negative the body floats upward at a velocity of -v.

However, a number of factors will lead to nonconformity with the equation:

- Most algae are not spherical. This effect is difficult to guantify, but in terms of settling, it may be expressed by a correctional term \emptyset_r , the coefficient of form resistance

$$\phi_r = \frac{v_s}{v'}$$
 (Eq 2)

anđ

$$\mathbf{v'} = \frac{2 \operatorname{gr}^2 (\rho' - \rho)}{9 \eta \cdot \phi_r}$$
 (Eq 3)

Suspended algal populations are usually dominated by live cells, whereas dead cells are rapidly eliminated. It has been demonstrated by a number of workers that dead diatoms, or even living senescent ones, sink faster than viable cells by factors 3 to 5 but without alteration in size or shape. For dead cells, there exist a strong correlation between sinking rate and cell radius, whereas for live cells there is no correlation. A comparison of measured v' and values of $v_{\rm c}$ calculated from Eq 1 for a number of freshwater algae showed that the sinking rate of most non-motile planktonic algae conforms in general to Stokes Law /Reynolds, 1984/.

In the marine environment, sinking rates of whole phytoplankton assemblages (not size-fractioned) range from $0.32 - 1.69 \text{ m d}^{-1}$; the average rate is $0.64 + 0.31 \text{ m d}^{-1}$ /Bienfang, 1981/.

Loss from the water column

"Loss" involves any process which actively removes biomass from the water body and depletes the stock of organisms. The loss processes affecting phytoplankton are hydraulic washout (i.e. the displacement of water supporting phytoplankton through an outflow), sedimentation, death and grazing. Sedimentation and grazing are considered the major factors governing the loss of phytoplankton from the water column.

Sedimentation

If the planktonic algae do not first decompose or if they are not consumed by animals they will inevitably settle onto the underlaying solid substratum. In a completely static water mass, a particle would sink at an average velocity of $v' m d^{-1}$ predicted by the modified Stokes equation (3). A water column of z m depth would then be progressively cleared of algal particles. If a large number of particles having identical sinking rates v' are initially distributed homogeneously through the water mass at a concentration of ${\rm N}_{\rm O}$ particles m⁻³, the individual particles will settle within the time range 0 - t', clearance being completed in t' = $\frac{z}{v}$, days. At any intermediate time \underline{t} the proportion of the original suspension which has settled approximates to $\frac{t}{t}$, or $\frac{\overline{v't}}{z}$.

The new concentration of particles is now

$$N_{t} = N_{0} (1 - \frac{t}{t})$$
 (Eq 4)

or

$$N_{t} = N_{0} (1 - \frac{v}{z} \cdot t)$$
 (Eq 5)

Assume then that the static water mass is instantaneously mixed such that the particles remaining in suspension are redistributed within the water column but that those having settled remain undisturbed and do no resuspend. If the number of mixings within the time interval 0-t' is \underline{m} , the mixing interval is $\frac{t'}{m}$. The amount of particles in suspension after the first mixing is then

$$N_{t} = N_{0} (1 - \frac{v'}{z} \cdot \frac{t'}{m})$$
 (Eq 6)

After the second, it will be

$$N_{t} = N_{0} (1 - \frac{v'}{z} \cdot \frac{t'}{m}) (1 - \frac{v'}{z} \cdot \frac{t'}{m}) (Eq 7)$$

and after the mth mixing

$$N_{t} = N_{0} \left(1 - \frac{v'}{z} \cdot \frac{t'}{m}\right)^{m}$$
 (Eq 8)

substituting t' = $\frac{z}{v}$, will give

$$N_{t} = N_{0} \left(1 - \frac{1}{m}\right)^{m}$$
 (Eq. 9)

Each period of quiescens thus reduces the amount in suspension to $(1 - \frac{1}{m})$ of its initial value. If this expression is expanded as a binomial series, it rapidly converges to a limiting value of

$$N_{t} = N_{0} \cdot \frac{1}{e}$$
 (Eq 10)

At fully developed turbulence when the particles still remaining in suspension are continuously mixed through the entire water mass, the deposition corresponds to a first order reaction. The fraction remaining at time t approximates

$$N_{t} = N_{0} \cdot e^{-k_{s} \cdot t}$$
 (Eq 11)

where $k_s =$ the loss rate constant (time⁻¹).

Reynolds /1979/ constructed a predictive model of sinking losses across the lower boundary of the mixed layer

$$\frac{dN}{dt} = -\frac{v'}{z_m}$$
(Eq 12)

where

N	=	the	mean population biomass within
		the	mixed layer
v	=	the	intrinsic sinking rate (m d^{-1})
^z m	=	the	vertical wind-mixed depth (m)

Equation 12 applies strictly to the dilution of a population of non-reproducing particles. Particles once lost from the mixed layer are thus destined to sink to the sediments. Reynolds /1979/ proposed that the sinking loss rate k_s can be approximated from v'/z_m. The population N_t remaining in suspension after t days is then given by

$$-\frac{v'}{z_{m}} \cdot t$$

$$N_{t} = N_{0} \cdot e \qquad (Eq 13)$$

The settling rate from static and from turbulent water, respectively, define the lower and upper limits of the sinking loss rate. The number of particles in suspension in a continuously-mixed suspension compared with the retention of the same particles from a static column of identical depth which is cleared completely in time t', is presented in Figure 1. For 99 % elimination, the period is approximately 4.6 times the minimum one predicted from the still-water velocity.

Grazing

Nanoplankton (size range 2 - 20 µ) consistently have high production rates considerably out of proportion to their total biomass /Kalff, 1972; Gelin, 1975/. Yet nanoplankton do not contribute overwhelmingly to the summer biomass in mesotrophic and eutrophic lakes. Thus, nanoplankton must suffer from high loss rates with grazing probably accounting for a large part of the loss. In fresh water, partitioning between the two loss processes, grazing and sedimentation, was found to vary interspecifically between almost complete removal (> 87 %) by grazers in the case of nanoplankton, and almost complete elimination (> 80 %) by sinking in the case of diatoms /Reynolds et al, 1982/.

Production and particle size

The production rates for the different size categories of natural phytoplankton crops are by no means similar. Observations by Kalff, /1972/ showed that organisms < 64 μ and < 20 μ contributed 75 - 79 % and 50 %, respectively, of the annual primary production. Netplankton (> 64 μ)

contributed more than half the biomass but never more than half the daily production. Similarly, Gelin /1975/ found that about 50 % of the annual production was synthesized by nanoplankton (< 20 µ) in a Swedish eutrophic lake. Besides, there was a 25-told variation of netplankton biomass throughout the year, compared with a 10-fold variation of nanoplankton biomass. The size-selective grazing by zooplankton is thought to be responsible for the dampened fluctuation in nanoplanktic abundance /Seliger et al, 1971/. Thus, the annual pelagic primary production in fresh water is characterized by a relative constancy in production by nanoplankton, compared to the more ephemerally significant contribution of netplankton of larger size.

Net change in standing stock

Despite the problems to develop theoretical models of the loss dynamics of natural populations, several attempts have been made to assess the different losses affecting phytoplankton populations in relation to changes in net standing stock. Knoechel and Kalff /1978/ tabulated cell growth rates $(0.08 - 0.41 d^{-1})$, death rates $(0 - 0.15 d^{-1})$, sinking rates $(0 - 0.48 d^{-1})$ and change in standing stock $((-0.41) - 0.38 d^{-1})$ for a number of diatom species over several periods during spring and summer. Similar findings were given by Reynolds et al /1972/ and Reynolds and Wiseman /1982/ which quantified the magnitude of losses to various sinks in relation to net standing stocks of diatoms. According to Reynolds and Wiseman /1982/, sedimentation accounts for differing proportions of the total loss of biomass for different algae: between 28

and 100 % of diatoms; 15 - 95 % of Eudorina (size \sim 100 μ) and less than 4 % of populations of small algae (<u>Ankyra</u>, <u>Chromulina</u>, <u>Cryptomonas</u>) (size \sim 15 μ). The authors postulated that other loss processes together accounted for 5 - 26 % of the observed maximal populations.

The loss rate of a spring diatom crop in a eutrophic lake, Northern Ireland, was assessed by Jewson et al /1981/. Net population growth was 4 % d⁻¹, washout 0.25 - 0.5 % d⁻¹ zooplankton grazing 0.05 - 0.15 % d⁻¹. A major decrease in the total phytoplankton population occurred during two very calm days when 50 % of one of the dominating species, Melosira italica (size $\sim 240 \mu$), settled out. However, it still took nearly a month before the crop was reduced to 5 % of its maximum. At the time of nutrient limitation, virtually the whole 90 % crop was reduced to 5 % of its maximum.

To sum up, the major processes affecting phytoplankton loss rates are sedimentation and grazing by zooplankton. Both processes are size-dependent; sedimentation is largely related to Stokes law, whereas grazing occurs to a greater extent within the nanoplankton (< 20 μ) size range. A continuously mixed water mass will extent the time required for particle clearance with a factor of about 5, compared with static conditions. As the adsorption of elements on particulate matter is size-dependent, the mechanisms affecting settling will also govern the loss of elements from the water column to the bottom sediments. In Figure 2 data are compiled concerning the size-dependent gains and losses of a hypothetical phytoplankton crop.

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Figure C-1 The number of particles retained in suspension in a continuously mixed suspension compared with the retention of the same particles from a static water column of identical height, which they clear completely in time t'.

Reference: Reynolds, 1984.



Figure C-2 Biomass and sinking rates of different size categories of phytoplankton.

References: Biomass data from Kalff, 1972 and sinking rates from Simpson, 1982.

APPENDIX D.

Distribution Coefficients in Soils

A literature survey on the sorption of iodine, radium, cesium, protoactinium, uranium, neptunium and plutonium on soils and clays is presented here.

Distribution coefficients are largely dependent on the pH, redox conditions, and complex forming ligands in the aqueous phase.

The redox conditions are only of interest for the actinide elements. The following predominance area diagrams illustrate the speciation for uranium, neptunium and plutonium at 25° C in waters containing 0.01 mol/l of carbonate (from Lemire-84 and Paquette and Lemire -81).





Table A.D. Kd values reported in the literature. Reference Iodine (I⁻) Sand (<2 mm) + Grndwater (pHz7) K_dz0.2-0.6 ml/g Hietanen-85 Soil $K_d \approx$ 1 to 50 l/kg Kocher-82 Fe_2O_3 +grndwater pH=3.5 K_d=0.1 m³/kg Andersson-82 Fe_2O_3 +grndwater pH=7-8 $K_d \approx 0.01 \text{ m}^3/\text{kg}$ Andersson-82 Montmorillonite+Grndwater pH=3-4 K_d=0.01 m³/kg Allard-80 " pH=7-8 K_d=0.0006 ** ** Al(OH)₃(c)+grndwater pH=7-8 K_d≈0.01 m³/kg Quartz+grndwater $pH=4 K_d=0.006; pH=7 K_d \approx 0.001$ ** Attapulgite+grndwater pH=4-5 K_d=0.06 m³/kg ... " pH=7-8 K_d≈0.01 ** Bentonite+grndwater pH=9 $K_d = 1.4$ m³/kg Torstenfelt-86a Radium (Ra²⁺) Sand K_d= 10 400 ml/g (100 to 38 000) Sheppard-84 $K_{d} = 300 \ 000 \ ml/g$ Silt ** K_d= 15 000 ml∕g (700 to 56 000) ** Clay Cesium (Cs⁺) Sand (<2 mm) + Grndwater (pHz7) K_dz1000 ml/g Hietanen-85 Clays+Grndwater (pH 7-8) K_d=200-18000 ml/g Miettinen-82 Bentonite+grndwater pH=9 $K_d = 1.4 \text{ m}^3/\text{kg}$ Torstenfelt-86a Protoactinium SiO_2 +0.01M NaClO₄ Pa(V) pH=3-4 K_d=0.2 m³/kg Allard-83 " [™] pH=7-8 K_d≈10 m³/kg ** $Al_2O_3+0.01M$ NaClO₄ Pa(V) pH=3-4 K_d=0.1 m³/kg Allard-83 " " pH=7-8 K_d≈10 m³/kg ... Bentonite+grndwater Pa(V) pH=9 $K_d = 5 m^3 / kg$ Torstenfelt-86b Uranium Attapulgite+grndwater U(VI) Andersson-82 pH=4-5 $K_{d}=0.8-10$ m^{3}/kg pH=7-8 $K_{d}=60-100$ m³/kg Montmorillonite+grndwater U(VI) Andersson-82 pH=4-5 K_d=2-10 m³/kg pH=7-8 K_d=1-5 m³/kg Allard-83 ** 11

Table A.D. Continued.

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Reference
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Neptunium	
Attapulgite+grndwater Np(V)	Andersson-82
pH=4-5 K _d ≈0.1 m ³ ∕kg	
pH=7-8 K _d ≈10 m ³ /kg	
Montmorillonite+grndwater Np(V)	Andersson-82
pH=7-8 K _d =0.1-0.05 m ³ ∕kg	
Al ₂ 0 ₃ +0.01 NaClO4 Np(V) pH=7-8 K _d =0.01-0.001 m ³ /kg	Allard-83
Bentonite+grndwater Np(V) pH=9 K _d =0.12 m ³ /kg	Torstenfelt-86 <u>b</u>
Clay+water pH=4-5 NpO ₂ ⁺ K _d =6 ml/g	Billon-82
pH=7 Np0 ₂ C0 ₃ K _d ≈100-10000 ml⁄g	**
Illite+0.1M NaCl Np(V) pH=5 K _d =8 l/kg	Meyer-85
" " pH=7-8 K _d =90-300 l/kg	Meyer-85
Boom Clay+Inters.sol. Np(OH) ₅ /Np0 ₂ CO ₃	
pH=8-9 K _d =130-1800 ml/g	Henrion-85
Seabed Sedim.+Sea water	
anoxic Np(OH) ₃ ' $K_{a} = 14000 - 67000 \text{ ml/g}$	Stanners-86
$0 \times 10 \text{ NpO}_2^{-1} \text{ K}_{d} = 1000$	
Sea sedim.+Sea water Np(V) K _d =1400-9500 ml/g	Higgo-83
Plutonium	
Attanulgitetgrndwater Pu(IV)	Andersson-82
nH=7-8 K = 60-100 m ³ /kg	
Montmorillonitetarndwater Pu(IV)	Andersson-82
nH=4-5 K =10 m ³ /kg	
pH=7-8 K = 20 m ³ /kg	
$Al_0 + 0.01$ NaClO4 Pu(IV)/Pu ³⁺ pH=2 K_=0.01 m ³ /kg	Allard-83
" " $Pu(IV) pH=7-8 K_{s}=5 m^{3}/kg$	11
Clav+water pH=7-8 Pu(IV) K_{230000} ml/g	Billon-82
" $pH=3-5$ Pu(VI) Kd ≈ 150 ml/g	11
" pH=7-8 Pu(VI) K_z=60000 ml/g	"
Boom Clay+Inters.sol. Pu(IV)/Pu(III)	
pH=8-9 K _a =160-10000	Henrion-85
Sea sedim.+Sea water Pu(V)/Pu(IV)≈0.5	
K _d =47000-95000 ml∕g	Higgo-85

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