Uranium and radium in Finnsjön – an experimental approach for calculation of transfer factors

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APPROACH FOR CALCULATION OF TRANSFER FACTORS

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Studsvik Energiteknik AB, 1981-05-07

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KBS 16.32

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ABSTRACT

The radiological safety studies for underground
disposal of HLW show that the future individual
and collective doses to an important extent may
originate from groundwater borne radium and
uranium which reach the biosphere. Indications
that the dispersion rates presently used give
rise to overestimations of calculated doses
justified an investigation for more realistic
turnover rates of radium and uranium than those
which now are in use.

Within one of the sites selected for testing,
the area around lake Finnsjön, a small number of
environmental samples were collected and analy-
zed with respect to radium and uranium and the
new transfer coefficients between soil and lake
water were derived.

The dose rates obtained with the new transfer
factors show a close agreement for radium and a
slight increase for uranium compared with ear-
lier calculations.

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1.   INTRODUCTION

The principal mechanism for the dispersion of
radioactive nuclides from HWL disposed under-
ground is the dissolution of the contaminants in
groundwater. The turnover rates of the nuclides
between groundwater and soil or lake or sea
water and bottom sediments are, however, hardly
known and data related to Swedish conditions in
particular are lacking. The on-going radiological
safety studies which are part of the Swedish KBS
program indicate that the potential future
individual and collective radiation doses to a
relatively important extent may originate from
groundwater borne radium and uranium which reach
the biosphere. The paucity of relevant information
contributes to the wide spans used in the calcu-
lated dispersion rates of radium and uranium in
those parts of the biosphere which are of impor-
tance when calculating the dose burden to man.

There are indications that the dispersion rates
which at present are used in the KBS studies can
give considerable overestimations of the calcu-
lated concentration in certain parts of the
ecosystem with respect to radium and uranium.
The apparent radiological importance of these
nuclides may therefore constitute an unrealistic
basis for site selection, design criteria and
safety evaluation.

To justify another choice of turnover rates than
those which now are in use, complementary infor-
mation are needed, preferably from the potential
disposal sites. Experimental efforts may improve
the calculations of the contributions to the
total dose burden from radium and uranium.

2.   DISTRIBUTION OF URANIUM AND RADIUM IN
            SOIL AND WATER

2.1   Migration of radium and uranium
            from bedrock deposits

Radium and uranium are present in the same kind of geological material. However, differences in geochemical behaviour with respect to weathering, sedimentation etc create a varied distribution of these elements in the environment. In decay chains with radioactive daughter products, the distribution of the daughter product also depends to a certain extent on the turnover process to which the parent nuclide is subjected. Uncertainties in the turnover of the parent nuclide can in some cases be amplified in the dose calculations for the daughter nuclide. Radium-226 with a half-life of $1.6 \cdot 10^3$ years is an alkaline earth metal with barium as nearest analogous and is a daughter nuclide of thorium-230 in the decay chain of uranium-234 - thorium-230 - radium-226.

The presence of radium and uranium in groundwater is dependent on their transfer rates from the bedrock to water and on their stability in solution. The transfers of radium and uranium are determined by the solubility of the bedrock which in turn is controlled by the chemical status of the water; oxygen and carbon dioxide tension, temperature etc (7). Oxygen and carbon dioxide are the gases which have the greatest influence on the mobility of radium and uranium, the solubility increases at low and high pH-values (4, 6, 18, 19). Diffusion processes between water and bedrock may promote an enrichment of minerals and alkaline and acid waters are usually enriched on radium and uranium.

When radium and uranium are transferred into
solution they are carried away with the ground-
water. The transport schedule is dependent of
the stability of the elements in the water as
well as the sorption capacities of the aquifer
rocks. The processes which can be responsible
for the precipitation of radium and uranium are
(6):

-       hydrolysis of uranium hydroxides
-       adsorption on surfaces of organic matter
-       breakdown of complex ions
-       reduction of the oxidation state
-       formation of insoluble salts

The interactions between groundwater and bedrock
are of special interest as governing mechanism
for the release of activity to the biosphere
from a hypothetical storage area for spent
nuclear fuel. In view of the relatively large
dose contributions involved, the decay chain of
uranium-234 - thorium-230 - radium-226 is of
particular interest. The amount of radium-226
appearing in the biosphere depends on how rapi-
dly uranium and radium are transported through
the soil, which influences the radium levels in
the groundwater.

Radium follows its analogous elements barium and
calcium. Adsorption processes are responsible
for the enrichment of radium in e.g. peat and
clay and radium is generally characterized by
significantly lower biological availability.
Table A.1 shows typical concentration values
for radium-226 in soil from different geogra-
phical areas.

2.2   Distribution of uranium and radium
          in the hydrosphere

The contents of uranium and radium in natural
waters are displayed in Table A.2. The average
concentration of uranium and radium in sea water
is 3 $\mu g \cdot l^{-1}$ and 0.1 $pg \cdot l^{-1}$ respectively. At
steady state conditions the ratio of radium to
uranium is the same as between their half-lives,
e.g. $3.6 \cdot 10^{-7}$. All values for radium/uranium
higher than $3.6 \cdot 10^{-7}$ indicate an accumulation of
radium to uranium. A deficit of radium relati-
vely to uranium exists in the oceans and to a
lesser degree in lakes. This is dependent on the
very low solubility of thorium-230 and its rapid
transfer to the bottoms through sedimentation.
This fact is supported by the excess of radium
to uranium that has been observed in the deep
sea sediments (8). Analysis of thorium-230 in
water and sediments have given a deficit in
water which is more marked than for radium.
Inversely, there is an excess of thorium-230 in
the sediments compated with Ra-226. This can be
explained by the fact that Th-230 accumulate in
the sediments where the daughter nuclide Ra-226
is generated and leached out to the overlaying
water (9, 10). Uranium and radium concentration
values for different fresh water localities in
Sweden are displayed in Table A.3, while
Table A.4 shows values of uranium and radium for
lake water and sediment from lake Hornborgasjön
and river Flian.

3.   DERIVATION OF TRANSFER COEFFICIENTS
BETWEEN SOIL AND LAKE WATER

Principles for derivation of transfer coefficients
from field measurements of concentrations in
different parts of a defined ecosystem are
displayed below.
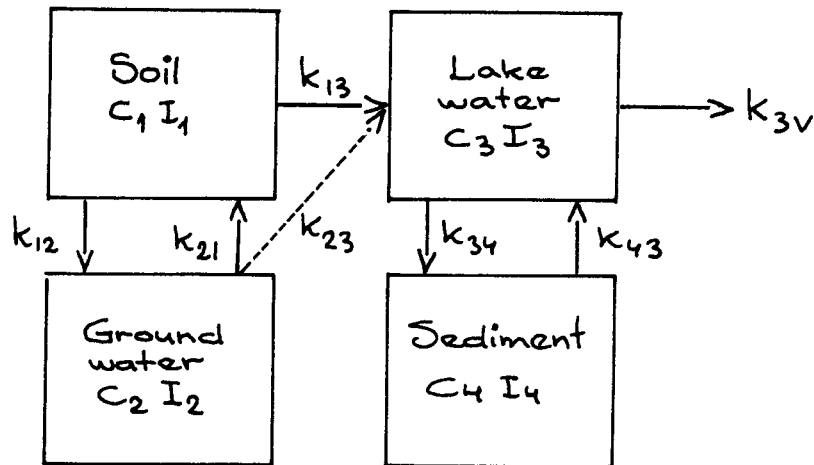
c_i = Concentration of reservoir $\underline{i}$

I_i = Inventory of reservoir $\underline{i}$

V = The discharge volume flow from the
          lake

$k_{i,j}$ = Fractional transfer rate from reservoir
          "i" to "j", i.e. the so called transfer
          coefficient

The transfer of uranium and radium from ground-
water to lake water is assumed to occur under
equilibrium conditions in the soil and groundwater
reservoir

$$\frac{k_{1,2}}{k_{2,1}} = \frac{I_2}{I_1}$$

The direct transport from groundwater to the lake may be included whereby the above equation is expressed as follows

$$\frac{k_{1,2}}{k_{2,1}+k_{2,3}} = \frac{I_2}{I_1}$$

Similar chemical equilibrium is assumed to occur between lake water and lake sediment as between the soil and groundwater reservoirs

$$\frac{k_{3,4}}{k_{4,3}} = \frac{I_4}{I_3}$$

Making it a condition that the amount of activity which is supported to the lake is balanced by the amount removed through runoff and radioactive decay in water column and bottom sediment, the leaching rate from soil to lake water becomes

$$k_{1,3} \cdot I_1 = k_{3,v} \cdot I_3 + \lambda \cdot I_3 + \lambda \cdot I_4$$

Substitution of

$$k_{3,v} \cdot I_3 = C_3 \cdot V$$

gives

$$k_{1,3} \quad \frac{C_3 \cdot V + \lambda(I_3 + I_4)}{I_1}$$

The amount of groundwater directly discharging into the lake is assumed to be of minor importance, making $k_{1,3} \gg k_{2,3}$.

3.1   Area description

At the request of KBS, the Swedish Geological
Survey, (SGU) has started a long-term project
aimed to clarify the geohydrological situation
of a test area close to the lake Finnsjön in
northern Uppland (1).

The investigation area is about 25 km^2 and is
limited by a water-shed. The test area is rela-
tively flat, rich in outcrops and often swampy.
Big areas of forests and agriculture have been
drained by ditches. The soil mainly consists of
moraine. The clay content varies between 1
and 4 % (1). In connection with test drillings,
chemical analysis of the bedrock have been
performed (2).

Lake Finnsjön which is connected to the investi-
gation area is a long and narrow lake. Depth
soundings have recorded a maximum depth of about
4 m. The lake can be characterised as oligotrophic
- dystrofic, i.e. with low fertility and brown
water. It is connected to a lake system which
extends in N-S direction and which discharges
towards north. The lake system is fed with water
from surrounding forests and bogs. The area
seemed suitable to test the applicability of the
approach described to calculate the transfer of
U and Ra within this defined ecosystem.

3.2   Environmental parameters

The following environmental parameters were
assumed to be relevant for the investigation
area.

Lake Finnsjön

Area 4.4 km^2.

Average depth 2 m.

Volume $8.8 \cdot 10^9$ l.

Of the discharge from the drainage area around
the lake system 80 % is supplied to the area
upstream lake Finnsjön. The average runoff from
Finnsjön is calculated to $1.5 \cdot 10^{10}$ l·yr^{-1}. The
nominal residence time for the water of lake
Finnsjön then becomes

$$\frac{8.8 \cdot 10^9}{1.5 \cdot 10^{10}} = 0.6 \text{ yr}$$

Lake sediment

The sediment area is assumed to be the same as
for the lake surface, i.e. 4.4 km^2. A 10 cm deep
sediment layer is assumed to take part actively
in the exchange process with the overlaying
water. The water content of the sediment is 55 %
and its density is calculated to 1.2, which will
give a total active sediment weight (dry weight)
of $2.4 \cdot 10^{11}$ g.

Drainage area

The runoff from surrounding drainage area is
calculated to 10 l·s^{-1} km^{-2}. A runoff from lake
Finnsjön of $1.5 \cdot 10^{10}$ l·yr^{-1} will give a drainage
area of 48 km^2.

Soil

A drainage area of 48 km^2 and an assumed depth
of 2 m will give a soil volume of $9.6 \cdot 10^7$ m^3.
The average density of 2.2 gives a total weight
of $2.1 \cdot 10^{14}$ g actively participating in the
exchange processes.

3.3   Sampling and analysis

During September 1979 a few samples were randomly
collected from the Finnsjön area:

- water from the lake
- bottom sediment
- water from a well
- soil samples

The water sample was collected from a boat. The
sediment sample was collected with an Ekman grab
which penetrated 10 cm down into the substrate.
Groundwater was collected from a well situated
about 100 m from the shoreline. Soil samples
were collected from the uppermost surface layer
and at 0.5 m depth.

To compare the concentration values in lake
Finnsjön with values obtained in the marine
environment, water and sediment samples from the
Baltic (Tvären Bay) were analyzed with respect
to uranium and radium.

The uranium contents of the different samples
were determined with delayed neutron technique
while the radium contents were analyzed radio-
chemically.

4.   CALCULATION OF TRANSFER RATES OF
        URANIUM AND RADIUM BETWEEN SOIL AND
        LAKE WATER

4.1   Uranium

The concentration values of uranium in the
different reservoirs of the ecosystem are
displayed in Figure B.1.

The calculated inventories of uranium in the
different reservoirs are displayed in Figure B.2.

Calculation of the transfer coefficient $k_{1,3}$
between soil and lake water using the parameters
displayed beneath:

$$
\begin{aligned}
C_3 &= 1.7 \cdot 10^{-2} \text{ Bq } l^{-1} \\
V &= 1.5 \cdot 10^{10} \text{ } l \cdot yr^{-1} \\
I_1 &= 8.88 \cdot 10^{13} \text{ Bq} \\
I_3 &= 1.52 \cdot 10^{8} \text{ Bq} \\
I_4 &= 1.48 \cdot 10^{10} \text{ Bq} \\
\lambda &= 1.55 \cdot 10^{-10} \text{ } yr^{-1}
\end{aligned}
$$

$$
k_{1,3} = \frac{C_3 V + \lambda (I_3 + I_4)}{I_1} = 2.9 \cdot 10^{-6} \text{ } yr^{-1}
$$

When chosing the environmental parameters, the
inventory in the soil reservoir may be regarded
as the one comprising the largest uncertainty.
Dependent whether one uses the concentration
value of surface soil or deep soil in the cal-
culation, the transfer coefficient will range
between $(3 - 5)10^{-6}$ yr^{-1}.

4.2   Radium

Concentration values and calculated inventories
of radium for the different reservoirs are
displayed in Figure B.3 and B.4.

For the water samples of Finnsjön and Baltic,
values below the detection limit were obtained.

Calculation of the transfer coefficient $k_{1,3}$
between soil and lake water using the parameters
displayed beneath:

$$
\begin{aligned}
C_3 &= < 6 \cdot 10^{-4} \text{ Bq } l^{-1} \\
V &= 1.5 \cdot 10^{10} \text{ } l \text{ yr}^{-1} \\
I_1 &= 1.07 \cdot 10^{13} \text{ Bq} \\
I_3 &= < 5.55 \cdot 10^{6} \text{ Bq} \\
I_4 &= 9.25 \cdot 10^{9} \text{ Bq} \\
\lambda &= 4 \cdot 10^{-4} \text{ yr}^{-1}
\end{aligned}
$$

$$
k_{1,3} = 1.3 \cdot 10^{-6} \text{ yr}^{-1}
$$

Dependent whether one uses the concentration
value of surface or deep soil in the calcula-
tion, the transfer coefficient will range be-
tween $(1 - 2)10^{-6} \text{ yr}^{-1}$.

4.3   Choice of environmental parameters

A sensitivity test will indicate how variations
of environmental parameters will have influence
on $k_{1,3}$. Using the set of parameters chosen in
the calculations of $k_{1,3}$ for uranium and radium,
as a starting point, and varying the parameters
one by one, will give the percentage deviation
of k from the original value (Table A.5 - A.6,
Figure B.5).

- A change of the lake volume does not affect the transfer coefficient.

- A change of the sediment volume affect $k_{1.3}$ (Ra) but not $k_{1.3}$ (U).

- $k_{1.3}$ is inversely proportional to the soil volume.

- $k_{1.3}$ is proportional to the outflow rate from the lake

- The concentration values for Ra in lake water and sediment will cause a change of k in a proportional manner, while the concentration of U in lake sediment does not affect k_U.

5.   CALCULATION OF GROUNDWATER TURNOVER
          RATES

The transfer of groundwater can be derived from
differences in nuclide concentrations in ground-
water and lake water. Assume the same amount of
the nuclides are brought to the lake as the
amount removed from it, i.e. steady state con-
ditions prevail:

$$V_{gw} \cdot a_{gw} \cdot k_{2,3} = V_1 \cdot a_1 \cdot k_{3,v}$$

$$k_{2,3} = \frac{V_1}{V_{gw}} \cdot \frac{a_1}{a_{gw}} \cdot k_{3,v} \quad yr^{-1}$$

V   = the volume of the ground water (gw)
            and lake reservoir (1)

a   = the specific activity of the ground
            water and lake water

With a ground water reservoir that comprises
20 % of the total volume of the soil reservoir
we become

$$V_{gw} = 0.2 \cdot 9.6 \cdot 10^{10} = 1.92 \cdot 10^{10} \; 1$$

Uranium values

With the same parameters used to calculate the
transfer coefficient $k_{1,3}$

V_1 $= 8.8 \cdot 10^9 \; 1$
a_{gw} $= 2.6 \cdot 10^{-2} \; Bq \; 1^{-1}$
a_1 $= 1.7 \cdot 10^{-2} \; Bq \; 1^{-1}$

$$k_{3,v} = \frac{1.5 \cdot 10^{10}}{8.8 \cdot 10^9} = 1.7 \; yr^{-1}$$

we arrive to

$$k_{2,3} = \frac{8.8 \cdot 10^9}{1.9 \cdot 10^{10}} \cdot \frac{1.7 \cdot 10^{-2}}{2.6 \cdot 10^{-2}} \cdot 1.7 = 0.51 \ yr^{-1}$$

The turnover of the ground water pool will then be about 2 yr.

Radium values

$$a_{gw} = 2.0 \cdot 10^{-3} \ Bq \ 1^{-1}$$
$$a_1 < 6.29 \cdot 10^{-4} \ bq \ 1^1$$

The same calculations will give

$$k_{2,3} = 0.3 \ yr^{-1}$$

Since a_1 is an upper limit value the same should hold for the calculated $k_{2,3}$ and the turnover rate of the ground water pool will then be > 4 yr.

Variatons of environmental parameters and anomalies in the uranium and radium concentration values within the investigation area will contribute to fluctuations in the calculated turnover rate of the ground water (Table A.7). To create a more solid foundation for the derivations of the k-factors will demand a larger sampling effort with a more detailed cover of the precipitation area.

6.   SIGNIFICANCE OF CALCULATED TRANSFER
            RATES FOR THE DOSE BURDEN TO MAN

The obtained transfer rates $(3 - 5) \cdot 10^{-6}$ yr^{-1}
and $(1 - 2) \cdot 10^{-6}$ yr^{-1} for uranium and radium
respectively between soil and lake water can be
compared with the values $1.2 \cdot 10^{-3}$ yr^{-1} and
$1.8 \cdot 10^{-4}$ yr^{-1} earlier used (3), where the trans-
fer rate of uranium has been derived from leaching
experiments in agriculture soils (4). Uptake of
uranium and radium in clover and leaching rates
in different soils were studied using uranium
concentrated soil residues which were mingled
with agriculture soils. Comparison was also made
with soils in which uranylnitratesolution was
added. The experiments from (4) showed an increased
transport in the acid leaching-water compared to
the uranyl-nitrate-solution with higher pH; the
leaching rate amounted to about 1 % per year for
a one m thick earth-layer, while the radium
content of the perkolated water was almost nil.
These experimental studies indicate that uranium
is leached considerably more rapidly through
Swedish moraine soils than has been assumed in
previous studies on vitrified wastes (17). The
field and laboratory studies which have been
carried out with respect to strontium and radium
indicate that radium is dispersed considerably
more slowly than strontium through soils under
widely varying conditions. In our previous
studies, however, it has been assumed that
radium is leached through the soil at the same
rate as strontium.

The transfer coefficients used in earlier investi-
gations together with the values obtained in
this study are displayed beneath

$k_{1.3}(U)$	$k_{1.3}(Ra)$	Ref
$1.2 \cdot 10^{-3}$	$1.8 \cdot 10^{-4}$	(3)
$(3 - 5) \cdot 10^{-6}$	$(1 - 2) \cdot 10^{-6}$	this study

It is generally agreed that low leach rates reduce the potential hazard. However, the composition of the leaching solution, flow rates, temperatures and other factors may vary significantly with the environment.

To test the radiological consequences for man when using the new transfer factors for the soil-water system in this study compared to (3), an analysis was performed with the BIOPATH program. Simulating a continous release of 10^{-6} Ci yr^{-1} ($3.7 \cdot 10^{4}$ Bq yr^{-1}) of uranium-234 into the ground water pool of the biosphere model (3) and changing the transfer factor between soil and lake water will give the dose rates displayed in Table A.8. The dose rates show a close agreement for radium and a slight increase of the regional dose for uranium, compared with (3). In the case of uranium the lower transfer factor will cause an accumulation in the soil compartment which will give rise to an increased dose rate. For radium the relatively rapid decay rate will counterbalance the increased accumulation in the soil, and the dose rate remains unaltered.

For radium-226 generated by uranium-234, the increased dose rates obtained reflects the slower transfer of uranium which causes an increased amount of uranium and consequently an increased amount of radium in the soil compartment. This means a more intense contact with the

groundwater and, as the drinking water constitutes
the principal part of the dose contribution, an
increased dose rate.

It can however be discussed whether the water
discharge to the lake from surrounding land
areas consists of surface runoff or if it mainly
originates from the groundwater pool. Table A.9
displays the obtained dose rates if the transfer
factor between groundwater and lake is changed
to the new ones for the soil-lake. We arrive to
larger differences in the calculated dose rates
than in previous case. This reflects the import-
ance of deeper knowledge concerning the transfer
of radionuclides in different ecological sub-
systems to calculate the dose burden to man.

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Table A.1

Typical concentrations of Ra-226 in soil excluding
high background areas. From (6)

Region	Bq g^{-1} (dry weight)	Ref
British Guiana	$(1.1-5.9) \cdot 10^{-2}$	13
Czechoslovakia	$3.7 \cdot 10^{-3} - 1.4 \cdot 10^{-1}$	14
Germany	$5.6 \cdot 10^{-3} - 4.8 \cdot 10^{-2}$	15
Ireland	$4.8 \cdot 10^{-2} - 1.1 \cdot 10^{-1}$	14
Italy	$2.7 \cdot 10^{-2} \pm 4.1 \cdot 10^{-3}$	16
United Kingdom	$3.0 \cdot 10^{-3} - 5.6 \cdot 10^{-2}$	14, 15
USA	$3.0 \cdot 10^{-2} - 1.0 \cdot 10^{-1}$	14
USSR	$3.7 \cdot 10^{-3} - 4.8 \cdot 10^{-2}$	14

Table A.2

Content of Uranium and Radium in Natural Waters[a]. From (6)

Type of Waters	Natural Conditions	Radium (g/liter)			Uranium (g/liter)			$\frac{Ra}{U}$
		min.	max.	mean	min.	max.	mean	
Surface Waters	Oceans and seas	8.0×10^{-14}	4.5×10^{-11}	1.0×10^{-13}	3.6×10^{-8}	5.0×10^{-6}	3.0×10^{-6}	3×10^{-8}
	Lakes	1.0×10^{-13}	8.0×10^{-12}	1.0×10^{-12}	2.0×10^{-7}	4.0×10^{-2}	8.0×10^{-6}	3×10^{-8}
	Rivers	1.0×10^{-13}	4.0×10^{-12}	2.0×10^{-13}	2.0×10^{-8}	5.0×10^{-5}	6.0×10^{-7}	3×10^{-7}
Waters of Sedimentary Rocks	Zone of intensive water circulation	1.0×10^{-13}	6.0×10^{-12}	2.0×10^{-12}	2.0×10^{-7}	8.0×10^{-6}	5.0×10^{-6}	5×10^{-7}
	Zone of highly impeded water circulation	1.0×10^{-11}	1.0×10^{-8}	3.0×10^{-10}	2.0×10^{-8}	6.0×10^{-6}	2.0×10^{-7}	1×10^{-3}
Waters of Magmatic Acid Rocks	Zone of intensive water circulation (waters of the weathering shell)	1.0×10^{-12}	7.0×10^{-12}	2.0×10^{-12}	2.0×10^{-7}	3.0×10^{-5}	7.0×10^{-6}	1×10^{-6}
	Zone of impeded water circulation (waters of deep tectonic fissures)	2.0×10^{-12}	9.0×10^{-12}	4.0×10^{-12}	2.0×10^{-7}	8.0×10^{-6}	4.0×10^{-6}	2×10^{-6}
Waters of Uranium Deposits	Zone of intensive water circulation (waters of the oxidation zone)	8.0×10^{-12}	2.0×10^{-9}	8.0×10^{-11}	5.0×10^{-5}	9.0×10^{-2}	6.0×10^{-4}	1×10^{-7}
	Zone of impeded water circulation (waters of the reduction zone)	1.0×10^{-11}	8.0×10^{-10}	6.0×10^{-11}	2.0×10^{-6}	3.0×10^{-5}	8.0×10^{-6}	1×10^{-5}

[a] Modification of table from A. N. Tokarev and A. V. Shcherbakov, Radiohydrogeology, Moscow (1956) AEC-tr-4100 (1960).

Table A.3

Uranium and radium concentration values for different fresh water
localities in Sweden. From (5)

Uranium

Water type	Locality	Concentration (Bq ℓ^{-1})	Ref
Surface water	Pösan	$2.8 \cdot 10^{-1}$	(5)
	Flian	$(1.6-2.1) \cdot 10^{-1}$	(5)
	Hornborgasjön	$(1.3-3.5) \cdot 10^{-2}$	(5)
River water		$(5-3.5) \cdot 10^{-2}$	(11)
Bog water	Masugnsbyn	$3.8 \cdot 10^{-1}$	(11)
Spring water	Northern Sweden	$(4.3-20) \cdot 10^{-3}$	(11)

Radium

Water type	Locality	Concentration (Bq ℓ^{-1})	Ref
Surface water	Flian	$(8.1-13) \cdot 10^{-3}$	(5)
	Hornborgasjön	$(5.9-8.1) \cdot 10^{-3}$	(5)
Drinking water		$(7.3-37) \cdot 10^{-3}$	(12)

Table A.4

Values of uranium and radium for lake water and
sediment from lake Hornborgasjön and river Flian
(Sweden). From (5).

Uranium

Water (Bq l^{-1})	Sediment (Bq kg^{-1})
$1.0 \cdot 10^{-1}$	$1.0 \cdot 10^{2}$
$2.2 \cdot 10^{-1}$	$6.3 \cdot 10^{1}$
$2.3 \cdot 10^{-1}$	$6.3 \cdot {}^{1}$
$1.4 \cdot 10^{-2}$	$6.3 \cdot 10^{1}$
$3.6 \cdot 10^{-2}$	$4.4 \cdot 10^{1}$
$1.6 \cdot 10^{-1}$	$7.0 \cdot 10^{1}$

Radium

Water (Bq l^{-1})	Sediment (Bq kg^{-1})
$1.3 \cdot 10^{-2}$	$1.0 \cdot 10^{2}$
$8.1 \cdot 10^{-3}$	$6.9 \cdot 10^{1}$
$8.1 \cdot 10^{-3}$	$4.8 \cdot 10^{1}$
$5.9 \cdot 10^{-3}$	$1.9 \cdot 10^{1}$
$1.2 \cdot 10^{-2}$	$2.2 \cdot 10^{1}$
$1.0 \cdot 10^{-2}$	$5.2 \cdot 10^{1}$

Table A.5

Percentage deviation of k_u when varying one environmental parameter from +50 % to -50 % by volume

	-50	-40	-30	-20	-10	±0	010	+20	+30	+40	+50 % deviation
lake volume	±0	±0	±0	±0	±0	±0	±0	±0	±0	±0	±0
sediment volume	±0	±0	±0	±0	±0	±0	±0	±0	±0	±0	±0
soil volume	+100	+67	+43	+25	+11	±0	-9	-17	-23	-29	-33
runoff	-50	-40	-30	-20	-10	±0	+10	+20	+30	+40	+50
concentration lake	-50	-40	-30	-20	-10	±0	+10	+20	+30	+40	+50
concentration sediment	±0	±0	±0	±0	±0	±0	±0	±0	±0	±0	±0
concentration soil	+100	+67	+43	+25	+11	±0	-9	-17	-23	-29	-33

Table A.6

Percentage deviation of k_{Ra} when varying one environmental parameter from + 50 % to -50 % by volume

	-50	-40	-30	-20	-10	±0	+10	+20	+30	+40	+50 % deviation
lake volume	±0	±0	±0	±0	±0	±0	±0	±0	±0	±0	±0
sediment volume	-15	-12	-9	-6	-3	±0	+3	+6	+9	+12	+15
soil volume	+100	+67	+43	+25	+11	±0	-9	-17	-23	-29	-33
runoff	-35	-28	-20	-14	-7	±0	+7	+14	+21	+28	+35
concentration lake	-37	-29	-20	-13	-8	±0	+8	+13	+20	+29	+37
concentration sediment	-15	-12	-9	-6	-3	±0	+3	+6	+19	+12	+5
concentration soil	+100	+67	+43	+25	+11	±0	-9	-17	-23	-29	-33

Table A.7

Percentage deviation of groundwater turnover rate when varying one environmental parameter from +50 % to -50 % by volume

	-50	-40	-30	-20	-10	±0	+10	+20	+30	+40	+50 % deviation
lake volume	-50	-40	-30	-20	-10	±0	+10	+20	+30	+40	+50
ground-water volume	+100	+67	+43	+25	+11	±0	-9	-17	-23	-29	-33
concentration lake (U)	-50	-40	-30	-20	-10	±0	+10	+20	+30	+40	+50

Table A.8

Dose rates obtained with the new set of transfer coefficients
between soil and lake water compared with (3)

Ra 226

$k_{1,3}$	maximum local individual dose rate	maximum regional individual dose rate	coll dose rate	ref
(yr^{-1})	$(Sv\ yr^{-1})$	$(Sv\ yr^{-1})$	$(manSv\ yr^{-1})$	
$1.8 \cdot 10^{-4}$	$2.1 \cdot 10^{-8}$	$1.7 \cdot 10^{-9}$	$4.5 \cdot 10^{-6}$	(3)
$1.0 \cdot 10^{-6}$	$2.1 \cdot 10^{-8}$	$1.8 \cdot 10^{-9}$	$4.7 \cdot 10^{-6}$	(this study)

U 234

$k_{1,3}$	maximum local individual dose rate	maximum regional individual dose rate	coll dose rate	reg
(yr^{-1})	$(Sv\ yr^{-1})$	$(Sv\ yr^{-1})$	$(manSv\ yr^{-1})$	
$1.2 \cdot 10^{-3}$	$6.0 \cdot 10^{-10}$	$3.4 \cdot 10^{-11}$	$2.8 \cdot 10^{-7}$	(3)
$3.0 \cdot 10^{-6}$	$6.0 \cdot 10^{-10}$	$5.8 \cdot 10^{-11}$	$3.4 \cdot 10^{-7}$	(this study)

Ra-226 generated by U-234

$k_{1,3}$	maximum local individual dose rate	maximum regional individual dose rate	coll dose rate	ref
(yr^{-1})	$(Sv\ yr^{-1})$	$(Sv\ yr^{-1})$	$(manSv\ yr^{-1})$	
$1.8 \cdot 10^{-4}$	$7.6 \cdot 10^{-12}$	$4.1 \cdot 10^{-10}$	$5.3 \cdot 10^{-6}$	(3)
$1.0 \cdot 10^{-6}$	$8.8 \cdot 10^{-12}$	$1.0 \cdot 10^{-9}$	$6.7 \cdot 10^{-6}$	(this study)

Table A.9

Dose rates obtained with the new set of transfer coefficients
between ground water and lake water compared with (3)

Ra 226

$k_{2,3}$	maximum local individual dose rate	maximum regional individual dose rate	coll dose rate	ref
(yr^{-1})	$(Sv\ yr^{-1})$	$(Sv\ yr^{-1})$	$(manSv\ yr^{-1})$	
$4.0 \cdot 10^{-4}$	$2.1 \cdot 10^{-8}$	$1.7 \cdot 10^{-9}$	$4.5 \cdot 10^{-6}$	(3)
$1.0 \cdot 10^{-6}$	$2.1 \cdot 10^{-8}$	$2.4 \cdot 10^{-9}$	$6.1 \cdot 10^{-6}$	(this study)

U 234

$k_{2,3}$	maximum local individual dose rate	maximum regional individual dose rate	coll dose rate	ref
(yr^{-1})	$(Sv\ yr^{-1})$	$(Sv\ yr^{-1})$	$(manSv\ yr^{-1})$	
$1.0 \cdot 10^{-3}$	$6.0 \cdot 10^{-10}$	$3.4 \cdot 10^{-11}$	$2.8 \cdot 10^{-7}$	(3)
$3.0 \cdot 10^{-6}$	$6.0 \cdot 10^{-10}$	$3.4 \cdot 10^{-10}$	$1.0 \cdot 10^{-6}$	(this study)

Ra-226 generated by U-234

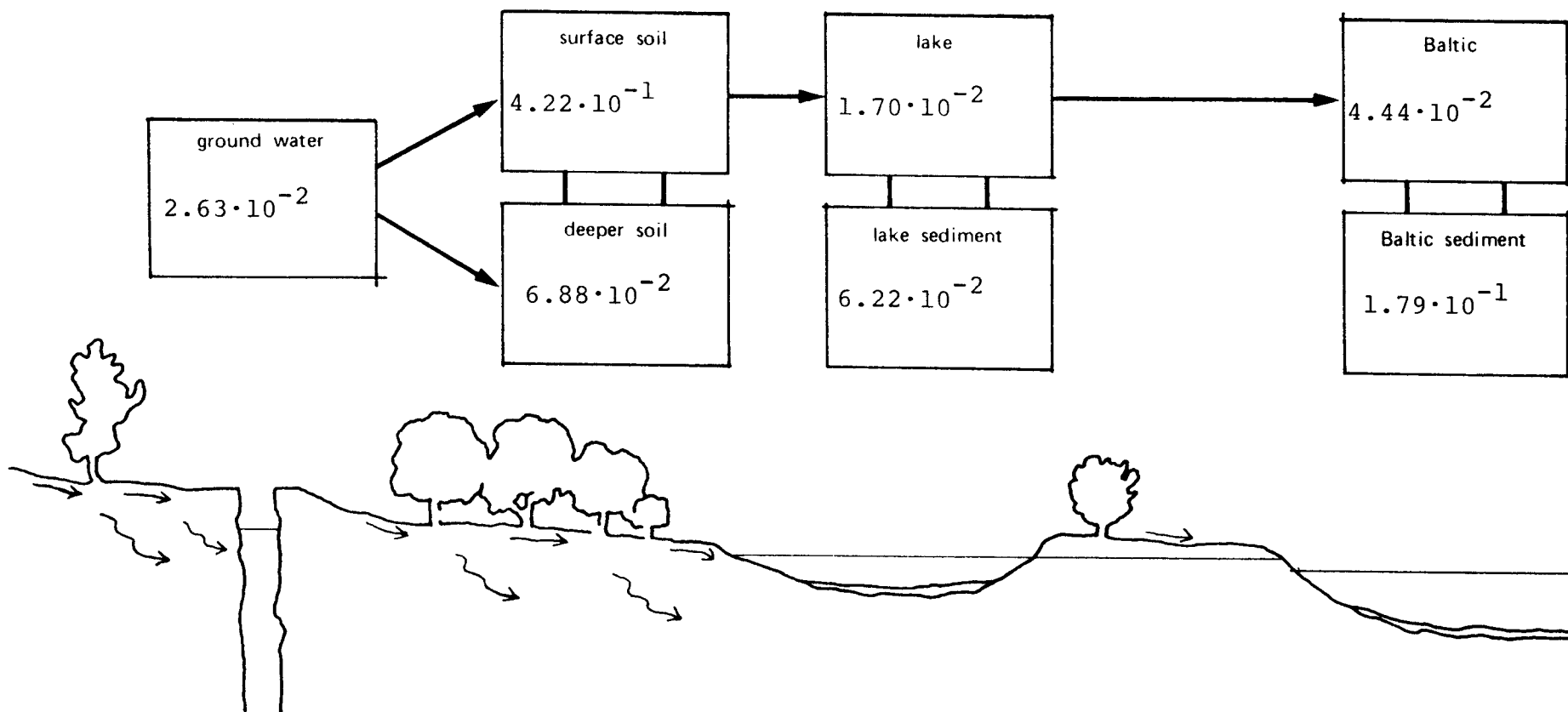| $k_{2,3}$	maximum local individual dose rate	maximum regional individual dose rate	coll dose rate	ref
(yr^{-1})	$(Sv\ yr^{-1})$	$(Sv\ yr^{-1})$	$(manSv\ yr^{-1})$	
$4.0 \cdot 10^{-4}$	$7.6 \cdot 10^{-12}$	$4.1 \cdot 10^{-10}$	$5.3 \cdot 10^{-6}$	(3)
$1.0 \cdot 10^{-6}$	$7.5 \cdot 10^{-12}$	$7.7 \cdot 10^{-9}$	$2.3 \cdot 10^{-5}$	(this study)

Figure B.1

Specific activity concentrations of U in the different reservoirs, expressed in Bq l^{-1} and Bq g^{-1} (dry weight) respectively.
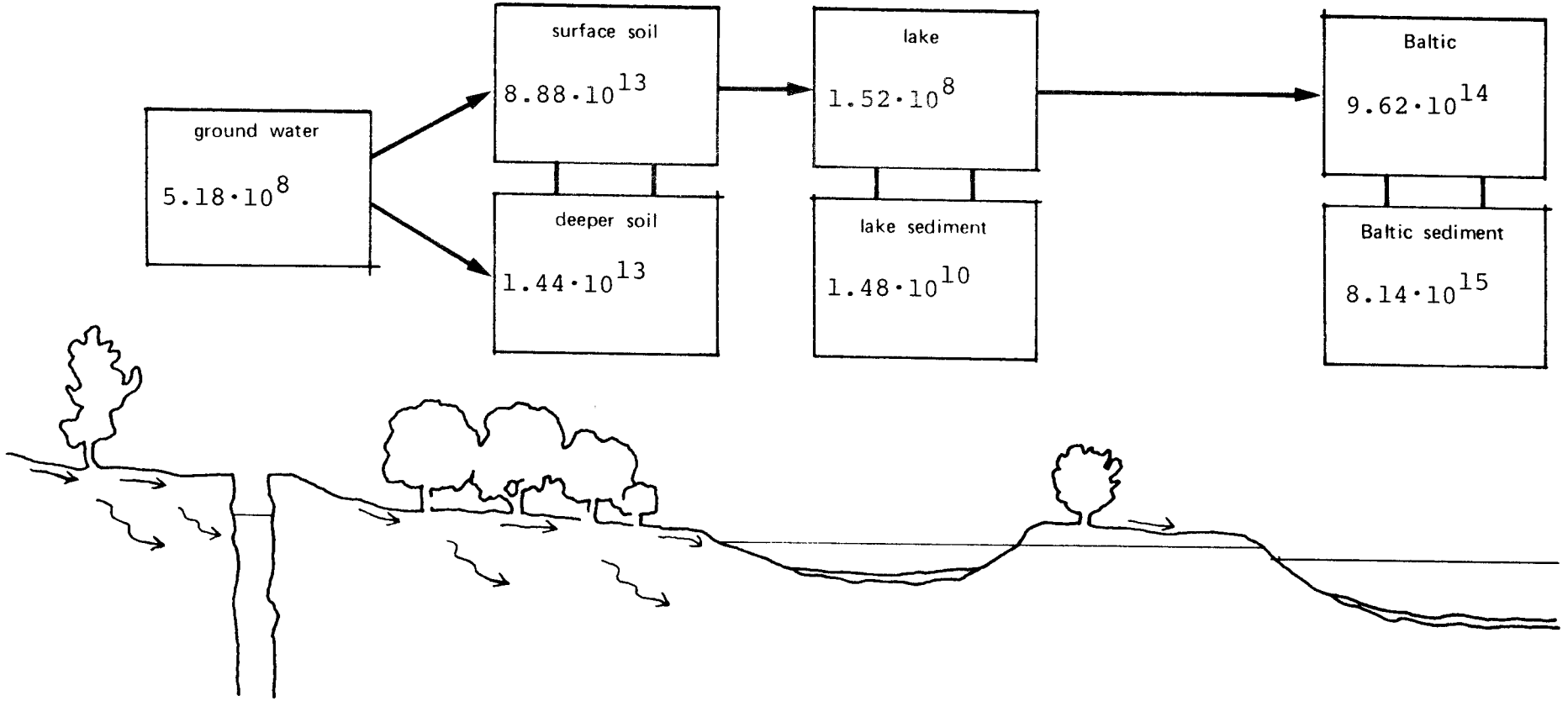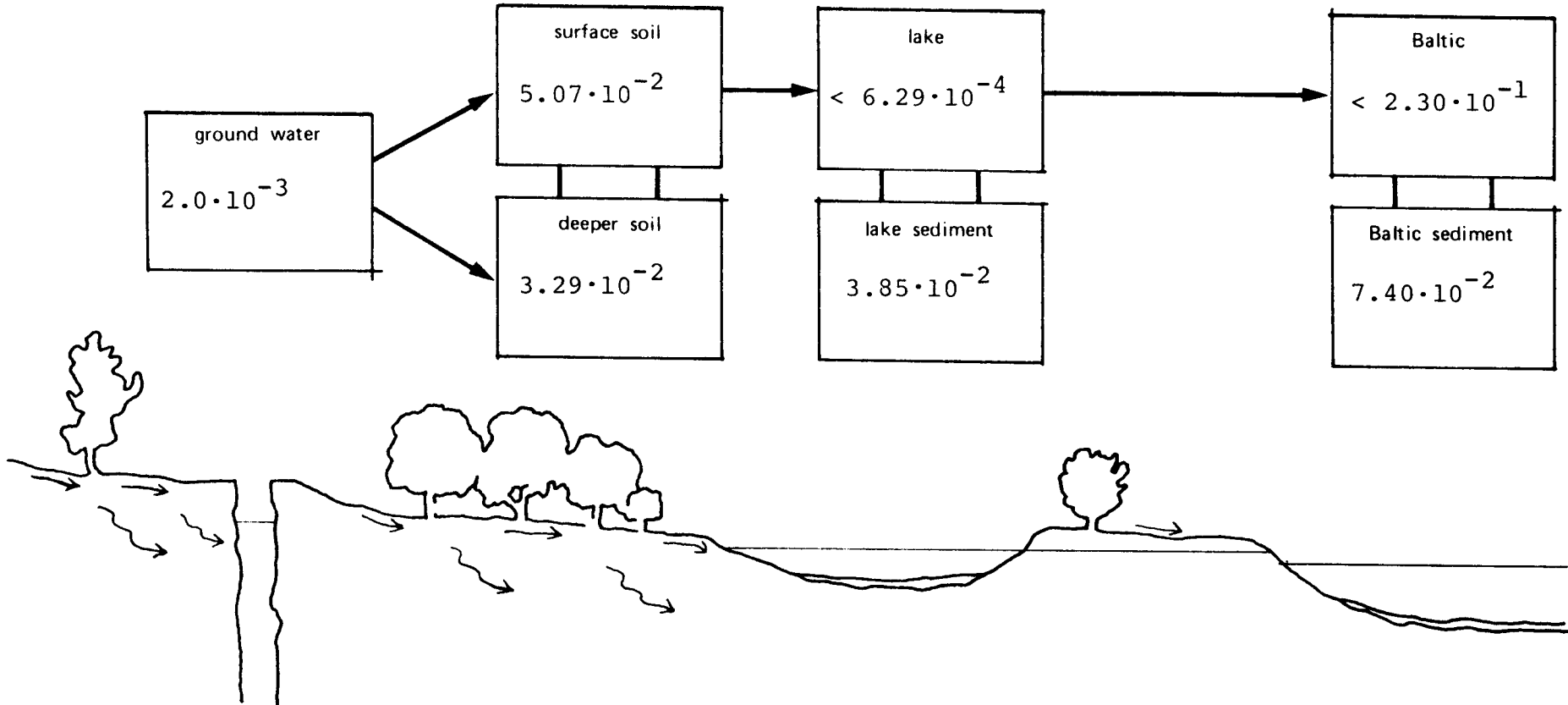
Figure B.2

Inventories of U in the different reservoirs expressed in Bq.

ground water	surface soil	lake	Baltic
$2.0 \cdot 10^{-3}$	$5.07 \cdot 10^{-2}$	$< 6.29 \cdot 10^{-4}$	$< 2.30 \cdot 10^{-1}$

deeper soil	lake sediment	Baltic sediment
$3.29 \cdot 10^{-2}$	$3.85 \cdot 10^{-2}$	$7.40 \cdot 10^{-2}$

Figure B.3

Specific activity concentrations of Ra in the different reservoirs, expressed in Bq l^{-1} and Bq g^{-1} (dry weight) respectively.
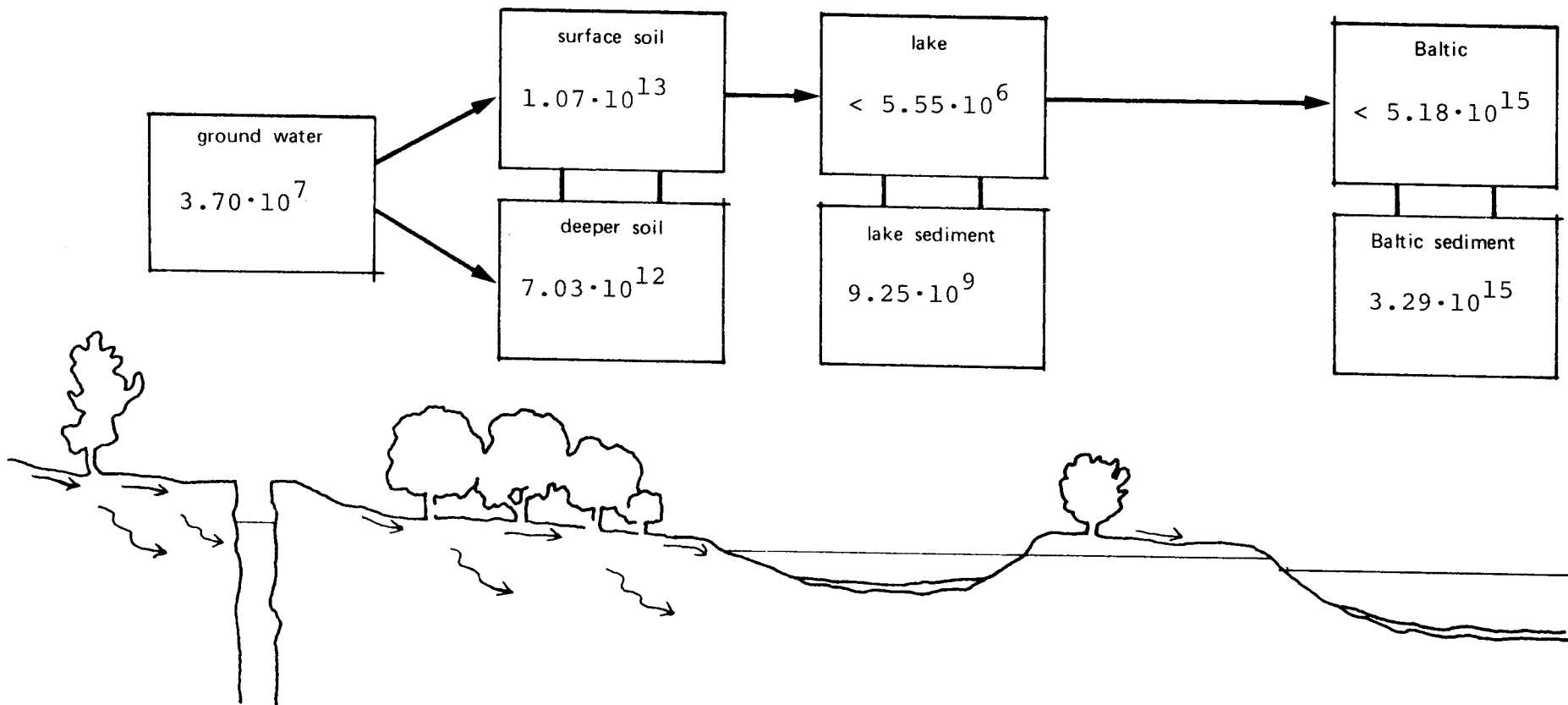
Figure B.4

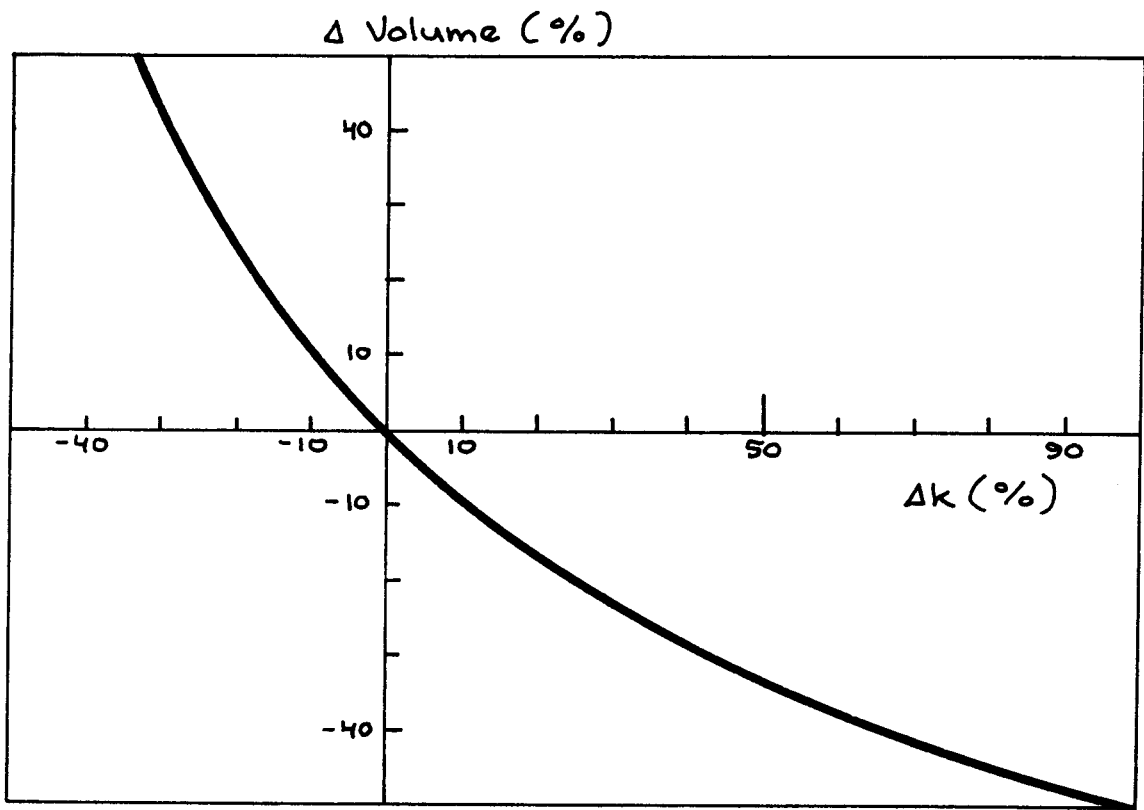Inventories of Ra in the different reservoirs expressed in Bq.

Δ Volume (%)

Δk (%)

Figure B.5

Percentage deviation of k_U and k_{Ra} with variation of the
soil volume.

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