



# **A model for the calculation of individual dose from exposure to radionuclides in contaminated soil.**

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The acceptable level of residual radioactivity in the ground at sites where radioactive materials have been handled is an important question for decommissioning. In order to answer this question, it is necessary to be able to estimate individual doses from exposure to radionuclides in the soil, taking the future use of the site into account.

This presentation gives an overview of a model which has been developed for the Swedish Radiation Safety Authority for the estimation of individual doses from exposure to radionuclides in contaminated soil. The model is generic, and calculates conservative estimates of individual doses intended for screening purposes. The results of the model, individual unit doses, form the basis for decisions about remediation and clearance of smaller areas of ground.

Dose calculations have been carried out for the radionuclides: H-3, Cl-36, Co-60, Ni-63, Sr-90, Tc-99, Cs-137, Np-237, Am-241, Pu-241, Pu-239, U-232 series, U-235 series and U-238 series, for times of up to 1000 years.

The calculated individual doses are based on assumptions about the exposure of people to soil which are, as far as possible, consistent with the assumptions used in the Swedish Environmental Protection Agency (SEPA) model for the calculation of generic guideline values for contaminated land.

## **Model description**

The SEPA model (SEPA, 2009) is used to calculate the concentration of contaminants in soil under which no harmful effects on health or on the environment are expected to occur. The guideline values are expressed as the total concentration of a contaminant in soil (mg/kg dry weight). The model developed for calculation of individual doses from radionuclides in soil only takes health risks into account. The model for radionuclides calculates the individual dose from a unit concentration of radionuclides in soil (1 Bq/kg).

## **Land use and exposure pathways**

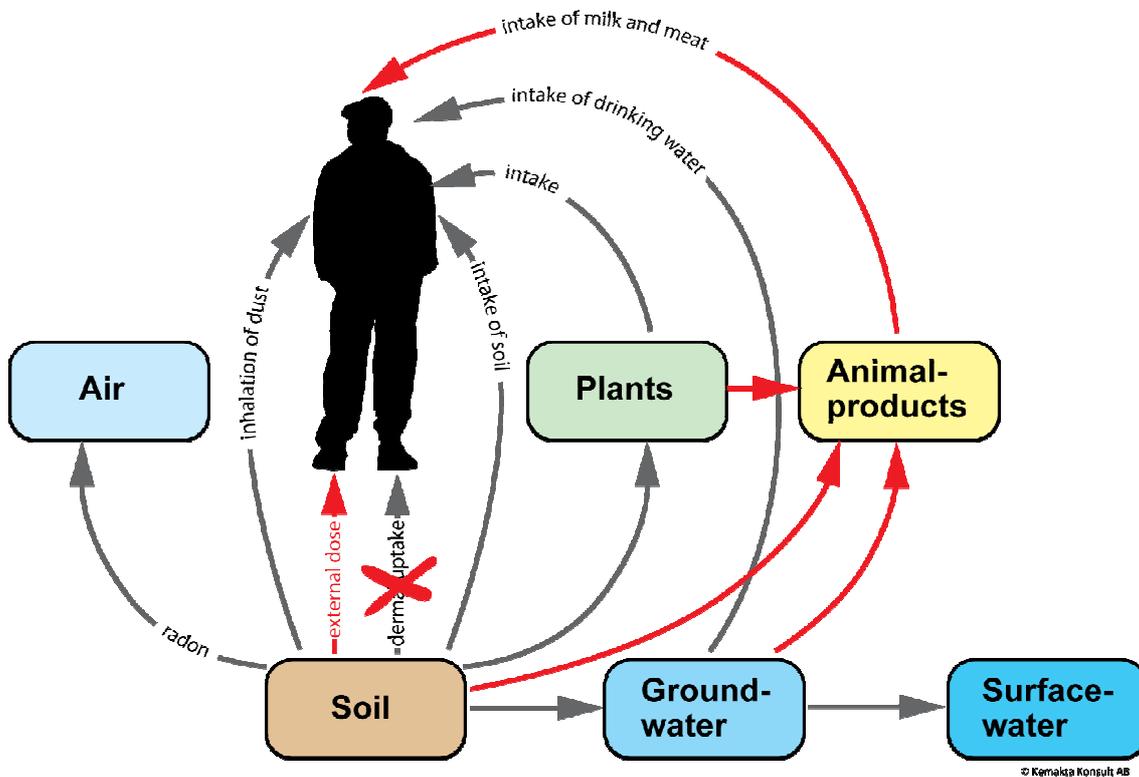
The contaminated area is assumed to be 50 m × 50 m, which is consistent with the SEPA model.

Two exposure scenarios for different types of land use are considered;

- Sensitive land use (e.g. housing, nurseries, schools, and parks) where the soil quality does not limit the land use, and groundwater can be used as drinking water. Full-time occupation of the contaminated area is assumed.
- Less-sensitive land use (e.g. industrial and commercial areas, roads and car parks) where the soil quality limits land use. The exposed groups are assumed to

be adults who occupy the contaminated area during working hours and children who visit the area only for limited periods.

The calculation of health risks from contaminated areas is based on the estimated exposure of people to contaminants in the soil. A number of exposure pathways are considered in the model, as shown in figure 1.



**Figure 1 Exposure pathways considered in the model (modifications to the SEPA model are indicated in red).**

The model for calculating individual doses from radionuclides was modified from the SEPA model with respect to the exposure pathways considered for the calculation of radionuclide-doses:

- Animal products (milk and meat) were included in the model. The animals are assumed to take in radionuclides in contaminated food and water, direct intake of contaminated soil and the inhalation of dust.
- External doses were included.
- The concentration of radionuclides in surface water is calculated, but exposure occurring from the use of surface water as drinking water or from the intake of fish is not included, as it is considered to be of less importance than the consumption of groundwater as drinking water.
- Exposure through the inhalation of contaminants in the gaseous phase is relevant only for radon and its daughter radionuclides. Scoping calculations have shown that the dose from the inhalation of tritium in the gaseous phase is of little importance compared to other exposure pathways. Therefore exposure through



inhalation has not been considered in the total individual dose from unit concentration in soil. For radon, the concentration of radon in indoor air is calculated and can then be compared with relevant criteria.

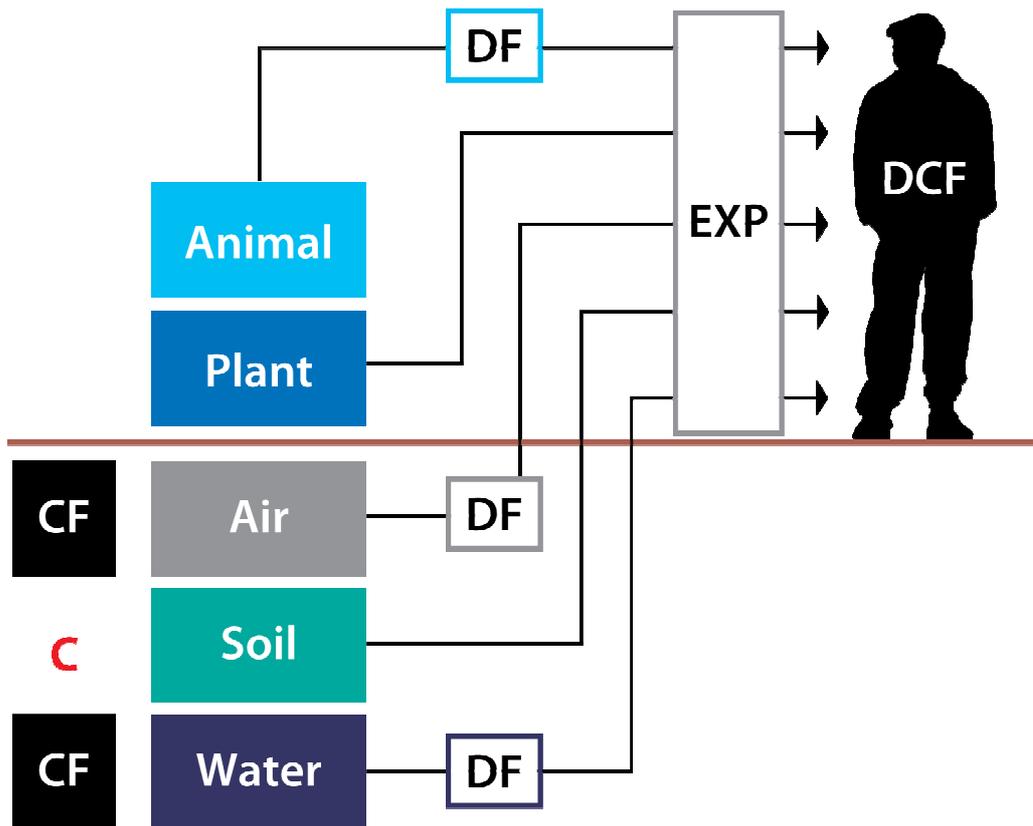
- Scoping calculations have shown that exposure through skin uptake and dose to the skin give a negligible contribution to the total dose. Therefore these exposure pathways have not been included.

<b>Exposure pathway</b>	<b>Sensitive land use</b>	<b>Less sensitive land use</b>
Direct intake of soil	Full time occupancy	Part-time occupancy
External dose	Full time occupancy	Part time occupancy
Inhalation of dust	Full time occupancy, indoor	Part time occupancy, indoor
Inhalation of radon gases	Indoor	Indoor
Consumption of vegetables grown on the contaminated area	10 % of total consumption of vegetables	Not considered
Consumption of drinking water	Well on the contaminated area	Not considered
Consumption of meat and milk	50% from animals that occupy the contaminated area	Not considered

### **Model structure**

The model is built up as shown in Figure 2 where:

- EXP     the mean yearly exposure for the radionuclide in the contact media (Bq/year)
- CF        the partitioning of the radionuclide between the soil and the contact medium (concentration in contact medium/concentration in soil, in simplified form)
- DF        the dilution between the contact medium before the contaminant reaches the exposed individual
- DCF      the dose conversion factor (Sv/Bq) for radionuclide intake by ingestion or inhalation, or (Sv/h)/(MeV·Bq/kg dry weight) for external dose.



**Figure 2 Description of the model for the calculation of individual doses.**

A radionuclide in soil with the concentration  $C$  is partitioned in soil (CF) between the different contact-media (soil, water, air, plant and animal products). For all pathways except those where exposure to the soil is direct, dilution occurs (DF) before the contaminant reaches the exposed individual. An individual is exposed for a certain amount of the radionuclide in the contact-media (EXP) and receives a dose which is calculated with the dose conversion factor.

Radioactive decay and the formation of daughter radionuclides are taken into account, which means that the inventory of radionuclides in the soil ( $C$ ) changes with time. However, the radionuclide inventory is not changed by the removal of radionuclides from the area by leaching with infiltrating rainwater, or other processes. This assumption is consistent with the SEPA model.

The concentration of dissolved radionuclides in the pore-water is assumed to be in equilibrium with the concentration of radionuclides sorbed to soil-solids, and is calculated from the total concentration of radionuclides using the equilibrium partitioning coefficient ( $K_d$ ) between soil solids and pore-water (mg/kg soil per mg/l pore water).

Radionuclides are assumed to be transported from the soil to groundwater, and to reach recipients in the form of a well or a surface water body (stream or lake). The contaminated pore-water is assumed to be diluted by uncontaminated water during transport from the contaminated area to the recipient.



The model which is used to calculate dilution during transport to a well is based on the following assumptions:

- When infiltration reaches the groundwater magazine, it is diluted by groundwater from areas upstream of the contaminated area.
- During transport between the contaminated area and the well, dilution occurs due to mixing in the sideways and vertical directions, as well as with rainwater infiltrating between the contaminated area and the well.
- The retention of radionuclides in the ground between the contaminated area and the well is not taken into account. Radioactive decay during the period of transport is not taken into account.
- The entire source area of the well lies within the contaminant plume.

Calculations with a model for the dilution of pore-water and data for typical Swedish conditions give a dilution-factor between pore-water and well-water of 14.

The model which calculates dilution during transport to a surface water body is based on the well-model, but also takes into account the dilution of the contaminated groundwater in the surface water. The contaminated groundwater is assumed to be mixed instantaneously in the entire volume of the surface water-body. This assumption is reasonable for small water-bodies. Calculations for typical Swedish conditions have been carried out and a dilution factor between pore-water and surface water was estimated to 4000 for a small lake or stream.

### **Sources of input data**

The generic guideline values are adapted for the environmental conditions usual at contaminated sites, and are calculated to protect health and the environment at the majority of contaminated sites in Sweden (though not all) by assuming reasonably conservative values for the model input parameters. This means that the calculated exposure can be higher than the average exposure on the area. However, unusual behavior, or other circumstances can lead to higher exposure than the calculated exposure, however, the probability of this is low.

The input data used in the model include:

- radionuclide specific data, such as decay half-lives, partitioning coefficients, dose-conversion factors and transfer factors,
- exposure factors, such as consumption habits and exposure times
- model parameters, such as the dilution factor pore-water to the groundwater aquifer used in estimation of radionuclide transport.

Radionuclide specific data were compiled for the model from literature reviews. Half-lives, mean energy per decay and decay fractions are well established and were taken from Firestone et.al. (1988) and ICRP (1983). Dose conversion factors were taken from EUs Basic Safety Standards (EG, 1996).

Partitioning coefficients for the distribution of radionuclides between soil-solids and pore water (K<sub>d</sub>-values) from a number of sources were compiled, including IAEA (1994) and IAEA (2005). For the metals Co, Ni and Pb, data from the SEPA model were used. The K<sub>d</sub>-values in IAEA (2005) are derived for modelling the leaching of radionuclides to groundwater and are generally lower than the values in IAEA (1994),



which are derived for modelling the retention of radionuclides in soils. For our model, which only uses Kd-values to model leaching behavior, the chosen Kd-values were in most cases based on IAEA (2005), although data from other sources were also taken into account in the choice of values.

Values used for the transfer factors to plant- and animal products were taken mainly from IAEA (2001). Plant uptake factors for Co, Ni and Pb were taken from the SEPA model. For some radionuclides, data were not available in IAEA (2001) and were taken from NCRP (1996). The transfer factors to meat and milk include the contribution of radionuclides from soil ingested together with the fodder by the animal, and are conservatively chosen.

The data for exposure factors and model parameters are consistent with the data used in the SEPA model, as far as possible. However an important adaptation made to the SEPA model is that individual doses from radionuclides are calculated using the available dose conversion factors for radionuclides (Sv/Bq) for six age-groups, whereas SEPAs model considers only two age-groups, adults and children (up to the age of 6 years). A number of exposure parameters are dependent on age-group, and therefore age-specific factors from Studsvik (2001) have been used for inhalation rate and the consumption of vegetables, meat, milk products and drinking water. For the sensitive land use scenario, the fraction of the total vegetable consumption grown on the contaminated area is assumed to be 0.1, based on data from the Netherlands (RIVM, 2001). All the drinking water consumed is assumed to come from the contaminated well.

The consumption rate of fodder and water by dairy and beef cattle has been taken from IAEA (1994). Because the contaminated area is assumed to be small, the fraction of cattle-fodder produced on the contaminated area was assumed to be 0.1. The fraction of the exposed individual's total consumption of milk- and meat which was assumed to come from animals consuming contaminated fodder was assumed to be 0.5. All the drinking water for cattle is assumed to come from the contaminated well.

### Decay series

The model includes radionuclides in four decay chains. For our calculations, the decay-chains were simplified. Daughter radionuclides with a short half-life in comparison with that of the parent radionuclide and with respect to the time period of interest for the calculations (1-1000 years), is assumed to be in equilibrium with the parent radionuclide. The dose from the short-lived daughter radionuclides is included in the dose calculations. An example is the U-238 series, see below, in which the radionuclides in bold type are included in the calculations of the decay series, but the radionuclides in normal type are assumed to be in equilibrium with their parent radionuclide.

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U-238	<b>U-238+</b> (Th-234, Pa-234m, Pa-234), <b>U-234, Th-230, Ra-226+</b> (Rn-222, Po-218, At-218, Pb-214, Bi-214, Po-214, Pb-210, Bi-210, Po-210)
U-234	<b>U-234, Th-230, Ra-226+</b> (Rn-222, Po-218, At-218, Pb-214, Bi-214, Po-214, Pb-210, Bi-210, Po-210)
Th-230	<b>Th-230, Ra-226+</b> (Rn-222, Po-218, At-218, Pb-214, Bi-214, Po-214, Pb-210, Bi-210, Po-210)
Ra-226	<b>Ra-226+</b> (Rn-222, Po-218, At-218, Pb-214, Bi-214, Po-214), <b>Pb-210+</b> (Bi-210, Po-210)
Pb-210	<b>Pb-210+</b> (Bi-210, Po-210)

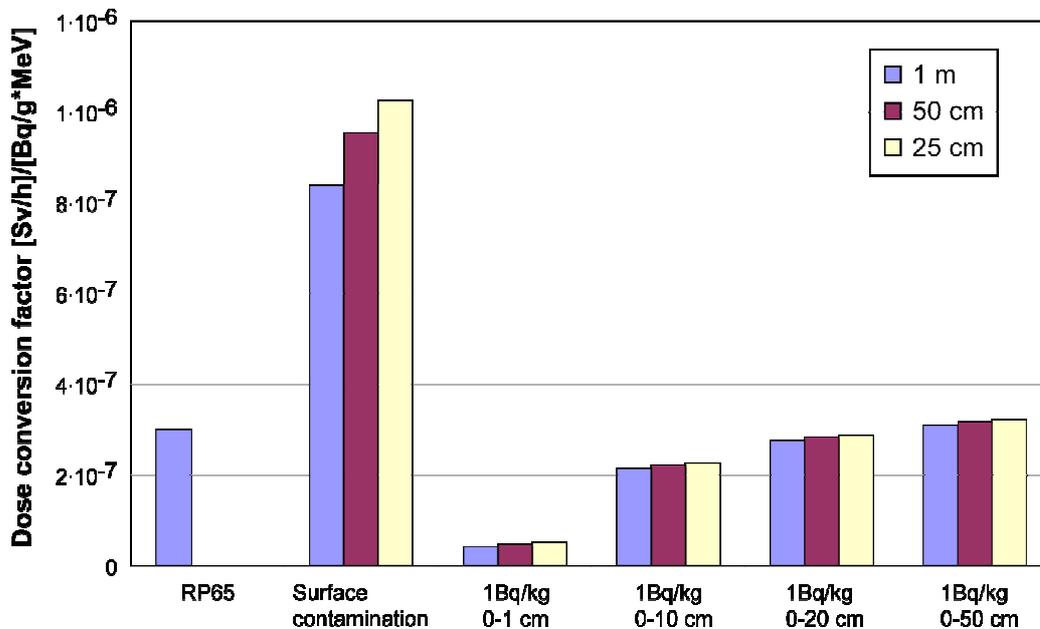
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An exception is made for the calculation of the dose from Ra-226 from the intake of drinking water, where radon-daughter radionuclides are not assumed to be in equilibrium (see Radon, below).

### External exposure

External dose is calculated using a dose conversion factor in Sv/h/(MeV·Bq/kg TS). The dose conversion factors are taken from RP 65 (EC, 1993).

Radionuclides are assumed to homogeneously distributed in the soil, even with depth. The effect of this assumption on the calculated external dose has been studied in a number of example calculations with the program “Microshield”. The dose rates at three heights above the soil surface has been simulated for soil with 1 Bq/kg of a radionuclide (with a decay energy between 0.01 and 5MeV) to a depth of 1, 10, 20 and 50 centimeters from the soil surface, as well as a case where all the activity was assumed to be at the soil surface (the same total activity as the case with 1 Bq/kg to a depth of 20 cm. See Figure 3.



**Figure 3 Dose conversion factors for external exposure at different heights above the ground surface for a radionuclide concentration of 1Bq/kg extending to different depths below the ground surface.**

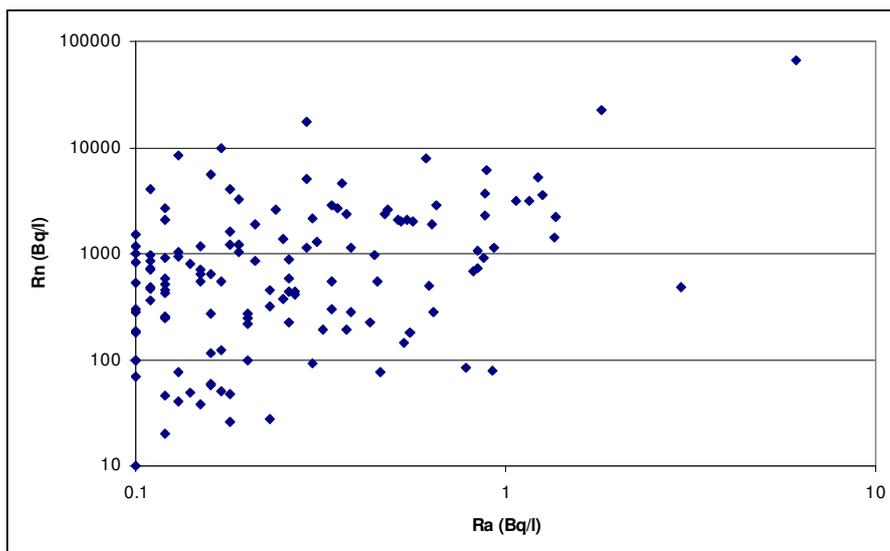
The results show that the dose rate increases with the contaminated depth, because the total activity increases. However, at depths below 10 cm, the increase in dose-rate is small because of the self-shielding of the soil. The calculated dose-rate for soil contaminated to depths of between 10 and 50 cm is similar to that used in report RP65, and therefore the dose conversion factor from RP65 has been used in model. If all the activity is at the soil surface, the model underestimates the external dose, but this is assumed to be unlikely.

## Radon

Dose from the inhalation of radon and radon daughters is not calculated in the model. Instead, the model calculates the concentration of radon in indoor air which can then be compared with the relevant guideline values and limits for radon in indoor air.

The concentration of radon in the air in soil pores can be calculated by assuming equilibrium with the radon concentration in the soil-air and soil-water phase using distribution coefficient, Henrys constant, of 4.4 (dimensionless). However, this gives an overestimation of the concentration of radon in soil air, according to measurements that SGU have carried out in Swedish soils (Jelenik 2008), probably because the model account for radionuclide decay, or the exchange of gases between the soil and the air above the soil. This approach is therefore not used in the model. Instead, the concentration of radon in the air in soil-pores is calculated using an empirical relationship between the radium concentration in soil and the radon concentration in soil air, which is based on SGUs data. The concentration of radon in indoor air is then calculated using the dilution factor for pore-air in indoor air from the SEPA model. The dilution factor of 6000 is based on a normally porous soil and is based on models developed to estimate the inflow of radon in soils into buildings.

The concentration of radon and radon daughters in groundwater is not in equilibrium with the concentration of radium in soil. This is because radon does not sorb to the soil solid phase, and is instead released into the water-phase. Measurements of the concentrations of radium, radon and radon daughters in a large number of Swedish wells (Ek et al. 2007) has shown that there is no clear relationship between the concentrations of radon and radium in wells. The ration Rn-222/Ra-226 varies between 100 and 100 000, with a median value of 5078 and a mean of 10 500, see Figure 4. When the data is restricted to wells with a radium concentration over 0.1 Bq/l, the ratio is somewhat lower, with a mean of 5860 and a median of 3000.



**Figure 4** The relationship between the concentration of Rn-222 and Ra-226 in 467 groundwater samples (Ek et. al. 2007).



The model calculates the concentration of radon in groundwater using a value of 1000 for the ratio Rn-222/Ra-226.

### **Tritium**

Because tritium, a radioactive isotope of hydrogen, occurs in the environment in many forms (including water and water vapour) and in biological materials, a large number of processes affect tritium transport and the exposure of people for tritium. There are different types of model to calculate doses from tritium in the environment. Specific activity models, based on the assumption that the ratio of tritium/stable hydrogen is the same in different parts of the environment, have been used by many organizations so estimate doses from tritium releases to the environment. Generally, specific activity models give a conservative assessment of doses, as they assume equilibrium between all environmental media, an unlimited, homogenous source of tritium and do not take into account the dilution occurring during radionuclide transport. Specific activity models are therefore less appropriate for the calculation of doses from a small area of contaminated soil. There are also models that account for the uptake of tritium into organic molecules where they are less exchangeable with stable hydrogen. These models do not assume equilibrium between the tritium activities in different hydrogen pools and require a large amount of site-specific data. These models are therefore less suitable for a generic screening model.

Our model calculates doses from tritium in the same ways as for other radionuclides. Tritium is assumed to be transported in the environment together with water, and has a very low  $K_d$ -value (effectively 0). Equilibrium concentration factors to calculate the uptake from soil to plant and the transfer from plants till animal products. The results of scoping calculations carried out by other organizations (Galeriu et al. 2007) with physiological models of tritium turnover agreed fairly well with the results of models using empirically-derived equilibrium transfer factors, and indicated that it is unlikely equilibrium models such as the one used in this study will underestimate the doses from tritium in soil.

Scoping calculations were carried out to estimate the contribution of the inhalation of tritium in indoor air to the total dose from tritium, using that the specific activity of hydrogen was in equilibrium in all model compartments, and assuming the same dilution of pore-air to indoor air as in the SEPA model. The inhalation of tritium in air was found to be of little significance compared with other exposure pathways, and has therefore not been included in the model.

### **Results**

The maximum total unit dose, which is obtained by adding the doses from the different exposure pathways, is shown for each radionuclide in Table 1. The doses shown in the table are the maximum dose for all age groups and times up to 1000 years.



**Table 1 The maximum total unit dose ( $\mu\text{Sv}/\text{year}$  per Bq/kg dry weight soil). The age group and the time for which the maximum dose is calculated is shown.**

Radionuclide	Sensitive land use			Less-sensitive land use		
	Max dose	Age group	time	Max dose	Age group	time
	( $\mu\text{Sv}/\text{year}$ )		(year)	( $\mu\text{Sv}/\text{year}$ )		(year)
Sum U-232	2.0E+00	12-17 year	10	6.8E-01	adult	10
Th-228+	1.0E+00	1-2 year	1	5.2E-01	adult	1
Sum Pu-241	2.2E-03	12-17 year	100	5.9E-04	adult	100
Am-241	7.7E-02	12-17 year	1	2.0E-02	adult	1
Sum U-238	1.1E-01	12-17 year	1000	1.4E-02	adult	1000
Sum U-234	1.1E-01	12-17 year	1000	2.9E-03	adult	1000
Sum Th-230	2.5E+00	12-17 year	1000	3.0E-01	adult	1000
Sum Ra-226	4.7E+00	12-17 year	100	8.2E-01	adult	1
Pb-210+	8.7E-01	1-2 year	1	5.9E-02	1-2year	1
Pu-239	4.7E-02	12-17 year	1	6.3E-03	adult	1
Sum U-235	2.7E-01	12-17 year	1000	9.2E-02	adult	1000
Sum Pa-231	1.3E+00	0-1year	300	2.5E-01	adult	300
Ac-227+	1.0E+00	0-1year	1	2.1E-01	adult	1
Co-60	2.0E+00	2-7year	1	1.1E+00	adult	1
Ni-63	3.6E-02	1-2year	1	4.0E-06	1-2year	1
Sr-90	1.6E+00	12-17 year	1	3.8E-04	12-17year	1
Cs-137	5.5E-01	12-17 year	1	2.6E-01	adult	1
H-3	1.2E-02	1-2year	1	2.2E-07	1-2year	1
Cl-36	3.7E+00	1-2year	1	7.8E-05	adult	1
Tc-99	1.3E+00	1-2year	1	2.3E-05	1-2year	1
Np-237	5.0E-01	0-1year	1	1.2E-01	adult	1

Figures 5 and 6 show the total dose for different radionuclides and age-groups for the time  $t=1$  year and the two different land uses. Radium has the highest unit dose for sensitive land use, for the age groups adults, 12-17 years, 7-12 years and 0-1 years. Cl-36 has the highest unit dose for 1-2 and 2-7 years. Co-60 has a consistently high dose for all age-groups.

For sensitive land use, the age-group 12-17 years has the highest unit dose for many of the radionuclides, and the younger age-groups have the highest dose for the radionuclides Co-60, Th-228, Ra-226, Ni-63, H-3, Cl-36, Tc-99, Ac-227 and Np-237.

For less sensitive land use, the highest unit for all age groups is calculated for Co-60, followed by Ra-226 and Th-228. The unit dose is greatest for adults for almost all radionuclides, except for Pb-210, Ni-63, Sr-90, tritium and Tc-99.

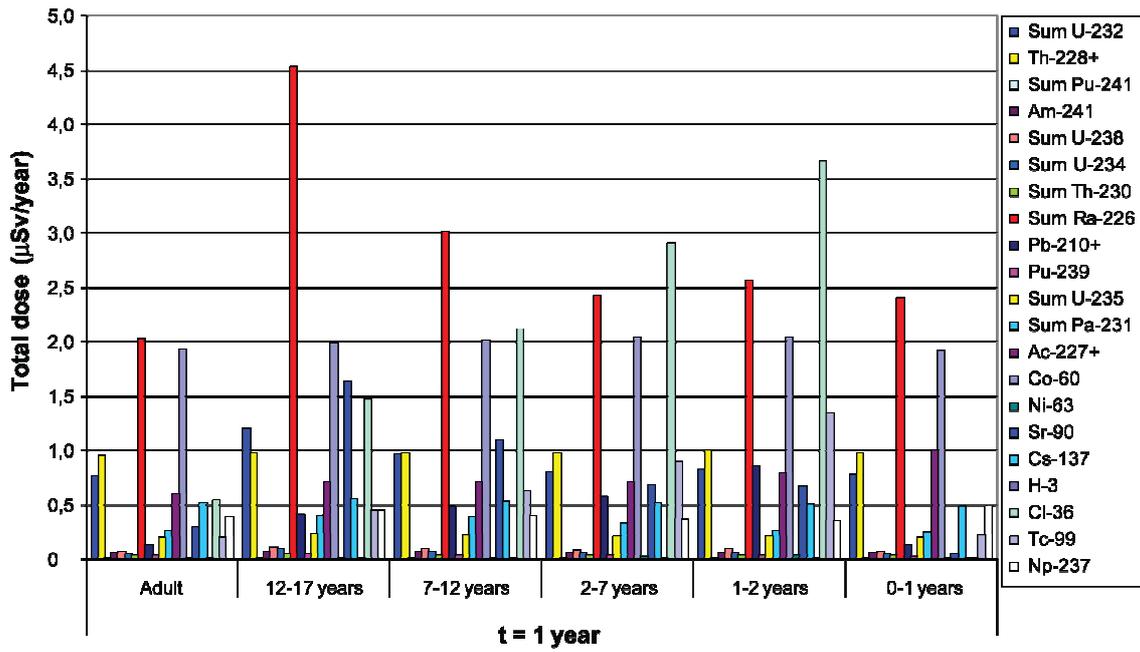


Figure 5 Unit doses from sensitive land use

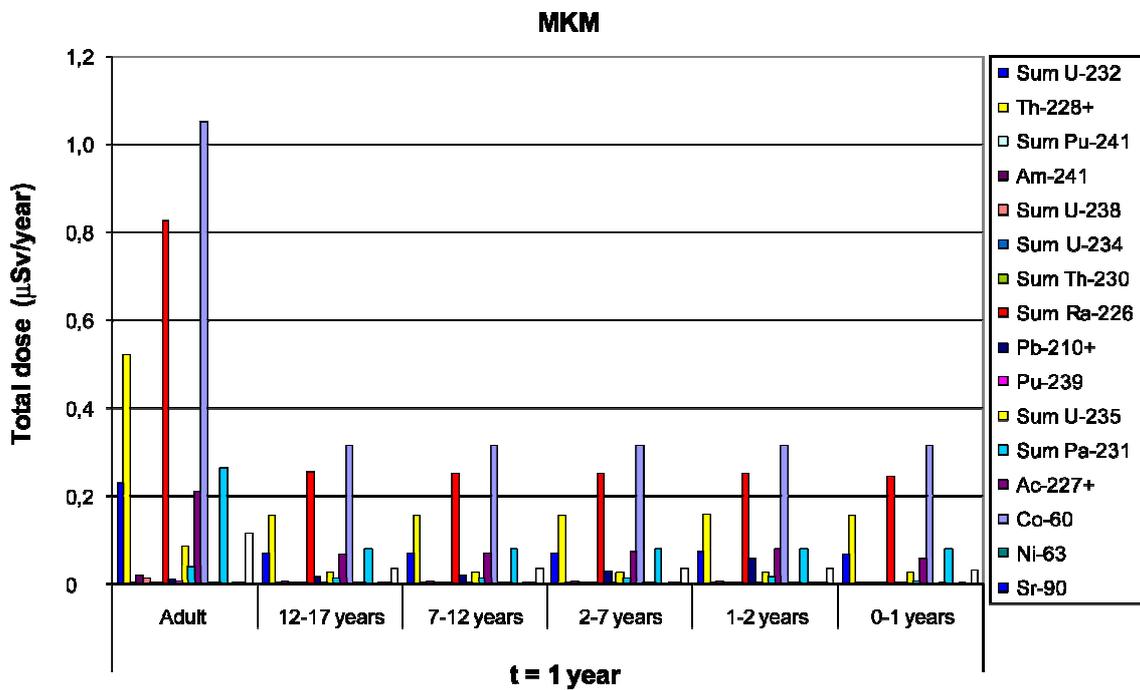
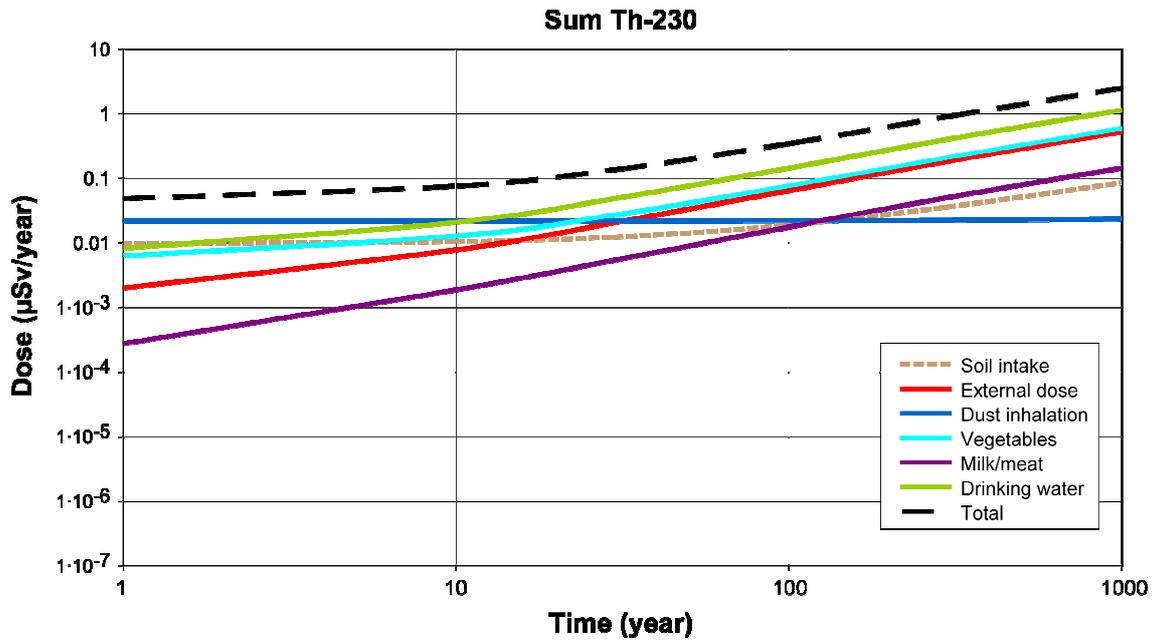
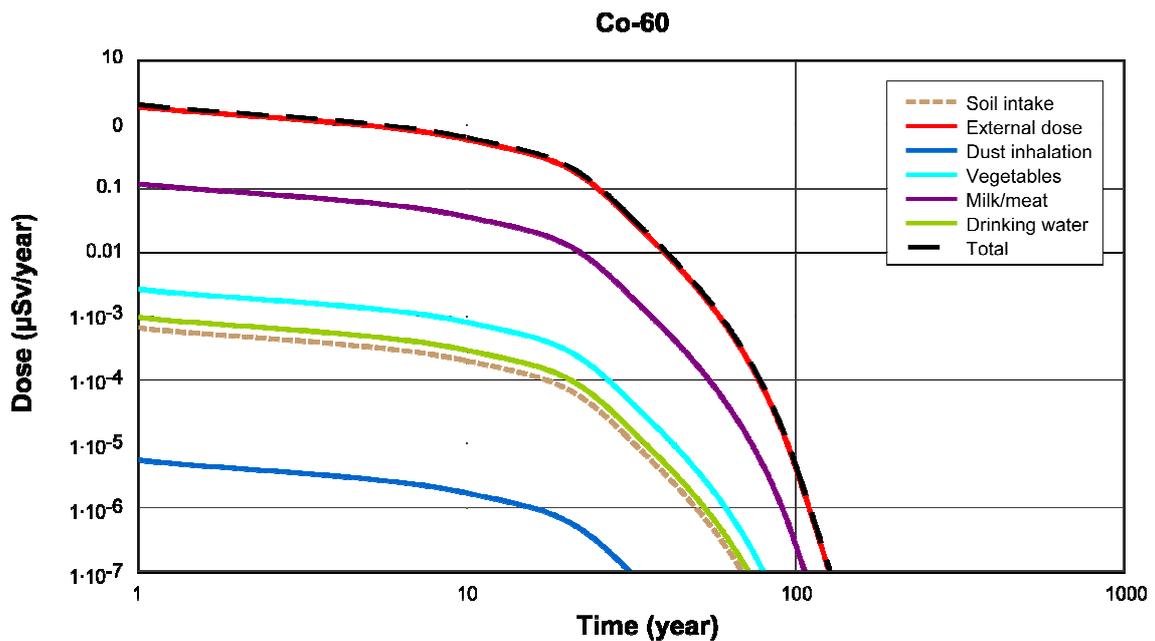


Figure 6 Unit dose, less sensitive land use

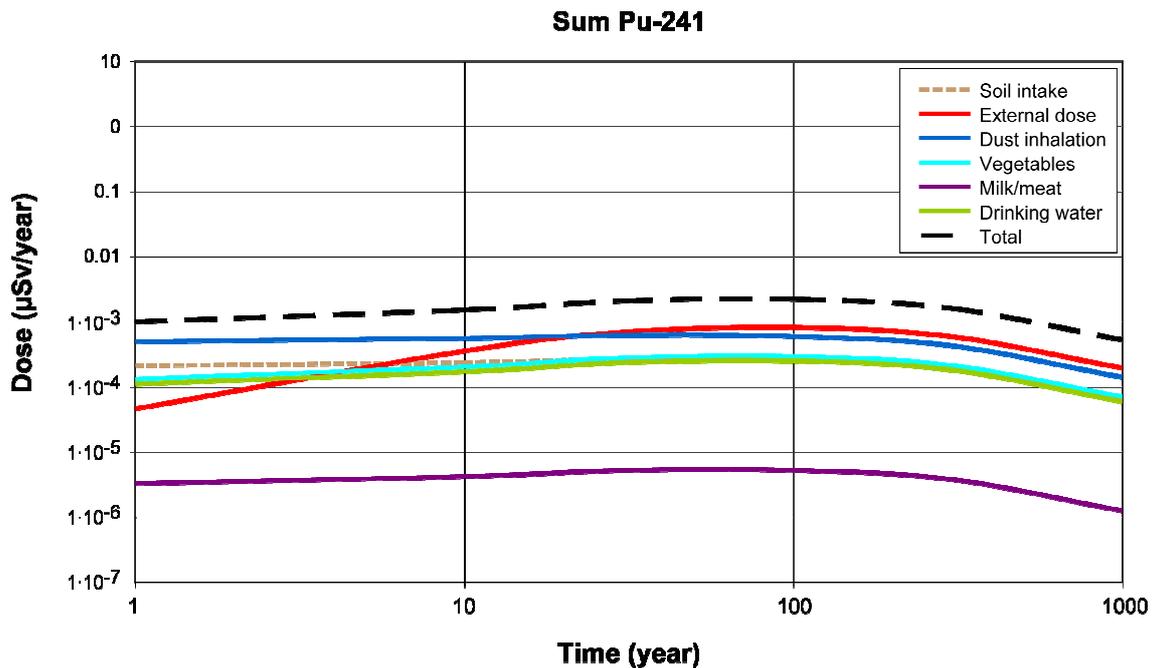
The change in unit doses with time can also be shown. Examples are shown in Figures 7 to 9 for Th-230, where the unit dose increases with time due to the ingrowth of the radionuclide, for Co-60 where the unit dose decreases due to radioactive decay, and for Pu-241, where the unit dose peaks during the 1000-year period.



**Figure 7** Dose from an initial inventory of Th-230 of 1 Bq/kg. Calculations for sensitive land use and age group 12-17 years.



**Figure 8** Dose from an initial inventory of Co-60 of 1 Bq/kg. Calculations for sensitive land use and age group 2-7 years.



**Figure 9** Dose from an initial inventory of Pu-241 of 1 Bq/kg. Calculations for sensitive land use and age group 12-17 years.

Tables 3 and 4 show the dominant exposure pathways for each radionuclide and the two land-use scenarios. For some radionuclides, the dominating exposure pathway changes with time (in which case, both pathways are shown in the tables). For less-sensitive land use, external dose dominates exposure for most radionuclides, at least during part of the time-period 0-1000 years. Exceptions are radionuclides where direct intake of soil is an important exposure pathway (and where the age-group with the highest unit dose is one of the younger age-groups).

External dose is the dominating exposure pathway even in sensitive land-use scenario for many radionuclides, at least during part of the time-period. The intake of animal products dominates for some radionuclides (Ni-63, Sr-90, H-3, Cl-36, Tc-99). The intake of drinking water dominates for U-232, U-238, U-234, Th-230, Pa-231, Ac-227, and Np-237. The inhalation is dust dominates for Pu-241 and Pu-239.

The dominating exposure pathways reflect the habits of the different age-groups: For the age-group 0-1 year, the unit dose is dominated by the drinking water pathway and external dose, as the consumption of animal products, vegetables, and the direct intake of soil are very low for this age group. For the age-group 1-2 years, direct intake of soil is more important than for other age-groups as hand-to-mouth activity is greater for this age group.

Sensitivity analysis has been carried out to investigate the effect of varying the following input parameters: K<sub>d</sub>-values, soil-to-plant transfer factors, the fraction of the total vegetable consumption which is grown on the contaminated area, and the fraction of the total amount of fodder and drinking water consumed by cattle that is derived from the contaminated area.



**Table 3 Dominating exposure pathways for each age group, sensitive land use**

Sensitive land use	Age group					
	Adult	12-17	7-12	2-7	1-2	0-1
Sum U-232	●	●●	●	●	●	●
Th-228+	●	●	●	●	●	●
Sum Pu-241	●●	●●	●●	●●	●●	●●
Am-241	●	●	●	●	●	●
Sum U-238	●	●	●	●	●	●
Sum U-234	●	●	●	●	●	●
Sum Th-230	●●	●●	●●	●●	●●	●
Sum Ra-226	●	●	●	●	●	●
Pb-210+	●	●	●	●	●	●
Pu-239	●	●	●	●	●	●
Sum U-235	●	●	●	●	●	●
Sum Pa-231	●●	●●	●●	●●	●●	●
Ac-227+	●	●	●	●	●	●
Co-60	●	●	●	●	●	●
Ni-63	○	○	○	○	○	●
Sr-90	○	○	○	○	○	●
Cs-137	●	●	●	●	●	●
H-3	●	○	○	○	○	●
Cl-36	○	○	○	○	○	●
Tc-99	●	○	○	○	○	●
Np-237	●	●	●	●	●	●

○ Dust inhalation	1	1	1	1	0	0
○ Milk/meat	3	5	5	5	5	0
● Drinking water	4	2	2	2	2	13
● External dose	9	7	9	9	9	7
● Vegetables	1	1	0	0	0	0
● Soil intake	0	1	1	1	2	0

**Table 4 Dominating exposure pathways for each age group, less sensitive land use**

Less sensitive land use	Age group					
	Adult	12-17	7-12	2-7	1-2	0-1
Sum U-232	●	●	●	●	●	●
Th-228+	●	●	●	●	●	●
Sum Pu-241	○ ●	○ ●	● ●	● ●	● ●	○ ●
Am-241	●	●	●	●	●	●
Sum U-238	●	●	●	●	●	●
Sum U-234	●	● ●	●	●	●	●
Sum Th-230	○ ●	○ ●	● ●	● ●	● ●	○ ●
Sum Ra-226	●	●	●	●	●	●
Pb-210+	●	●	●	●	●	●
Pu-239	○	○	●	●	●	○
Sum U-235	●	●	●	●	●	●
Sum Pa-231	●	●	●	●	●	●
Ac-227+	●	●	●	●	●	●
Co-60	●	●	●	●	●	●
Ni-63	●	●	●	●	●	○
Sr-90	●	●	●	●	●	○
Cs-137	●	●	●	●	●	●
H-3	●	●	●	●	●	○
Cl-36	●	●	●	●	●	●
Tc-99	●	●	●	●	●	○
Np-237	●	●	●	●	●	●

- Dust inhalation
- Milk/meat
- Drinking water
- External dose
- Vegetables
- Soil intake

## References

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