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Models used in the SFR 1 SAR-08 and KBS-3H safety assessments for calculation of ¹⁴C doses

Rodolfo Avila, Facilia AB

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Preface

The work presented in this report is a result of a project commissioned by Svensk Kärnbränslehantering (SKB) and Posiva Oy, with Anna Gordon and Ari Ikonen as the project coordinators, respectively. The report is printed and available also as a SKB report R-08-16/ Posiva Working report 2007-107.

This document presents a set of simplified models for assessment of human exposures resulting from potential underground releases of ¹⁴C. These models were used in the SFR 1 SAR-08 and KBS-3H safety assessments. The document constitutes one of the references used in the safety analysis SFR 1 SAR-08.

Rodolfo Avila, Facilia AB, Gerhard Proehl, ConRad- Consulting in Radioecology, Mueller und Proehl GbR have compiled the report.

This document has been reviewed and all comments have been documented in accordance with SKIFS 2004:1.

Stockholm, February 2008

Anna Gordon

Project leader, SFR 1 SAR-08

Abstract

This report presents a set of simplified models for assessment of human exposures resulting from potential underground releases of ¹⁴C. These models were used in the SFR 1 SAR-08 and KBS-3H safety assessments. The proposed models can be used to assess continuous, as well as pulse-like ¹⁴C releases, to various types of biosphere objects: forest ecosystems, agricultural lands, sea basins and lakes. It is also possible to make assessments of exposures resulting from the use of contaminated fresh waters, for example from an impacted well, for irrigation of vegetables. Models are also proposed for scenarios where lakes and sea basins are transformed into terrestrial objects due to land rise, filling of lakes and other natural or human induced processes. The exposure pathways considered in dose calculations with the models are: ingestion of contaminated food and water for both terrestrial and aquatic ecosystems, inhalation of contaminated air for terrestrial ecosystems. The exposure by external irradiation is not considered, as ¹⁴C is a pure low-energy beta emitter.

The report provides an overview of the behaviour of ¹⁴C in the environment, including an outline of the conceptual assumptions implicit in the proposed models. The proposed models are based on the so-called specific activity approach, which has been recommended by the UNSCEAR and the IAEA for assessment of doses resulting from ¹⁴C releases to the environment from nuclear installations, such as nuclear power plants. The equations for estimation of the ¹⁴C specific activities in environmental compartments have been derived from a combination of several realistic and conservative assumptions, which are documented and justified in the report. The models can be used in safety assessments of geological repositories of radioactive waste, to carry out cautious, but still not over conservative dose estimations, which can be compared with regulatory dose constrains.

Comparative studies with the models indicate that the worse case situations will be associated with releases to small lakes, especially if these have a low productivity and relatively long water residence times. Releases to terrestrial ecosystems and sea basins will generally result in lower annual doses per unit release rate, as in these systems the residence time in air and water, respectively, of the released ¹⁴C are very short compared with the characteristic times of assimilation of carbon by primary producers. The study also indicates that accumulation of ¹⁴C in lake and sediments and subsequent exposure to terrestrial systems formed on these sediments does not result in substantial annual doses per unit release rate, as compare with those received from releases to the corresponding aquatic ecosystem. In all cases considered in the study, the annual doses per unit release rate via food ingestion were much higher than the annual doses by other pathways, like inhalation and water ingestion, which is consistent with results reported from other studies.

The uncertainty analysis shows that in most cases the dose predictions are within a factor from 5 to 20. This is a smaller range of uncertainty than the ranges that are obtained in assessments using transfer factors, which show a variability of several orders of magnitude. The results of the sensitivity analysis indicate that few parameters have a dominant effect on the dose predictions. These are parameters that can relatively easily be measured, such as the wind speed, the concentration in water of dissolved inorganic carbon, the water residence time and the net primary production in the ecosystem.

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1 Introduction

Radiocarbon (¹⁴C) may be released from radioactive waste disposal facilities and cause exposures to man after entering the biosphere. Possible biosphere receptors for ¹⁴C ascending from a geological repository are terrestrial ecosystems like forests and agricultural lands, as well as water bodies such as wells, lakes, rivers and sea basins. Previous safety assessments of geological repositories in Finland and Sweden has shown that ¹⁴C could have an important contribution to doses to potentially exposed individuals, specially for the case of repositories for low and intermediate level radioactive waste /Lindgren et al. 2001/.

In assessments of radiological impacts from ¹⁴C releases from nuclear facilities, such as nuclear power plants, it is a common practice /IAEA 2001/ that special radioecological models, the so-called specific activity models, are used for this radionuclide, as well as for H-3. This is motivated by the strong influence that the carbon and water cycles respectively have on the environmental behaviour of these radionuclides. This was already recognised in the previous assessment of the SFR 1 repository, where a dynamic model /Kumblad et al. 2003/ was used to simulate the fate ¹⁴C releases to the marine environment. However, for releases to terrestrial and limnic environments the same type of models as for other radionuclides were used. The pitfalls of applying the same approach for ${}^{14}C$ as for other radionuclides have been acknowledged – see for example a discussion in /Avila et al. 2006/. Among other problems, the difficulties in finding appropriate values for ¹⁴C of transfer factors from soil or water to biota, and of distribution coefficients between solids and liquids, have been highlighted. Moreover, it has been questioned /Avila et al. 2006/ whether at all it is conceptually sound to use transfer factors and distribution coefficients for ¹⁴C, as these usually do not reflect the actual transfer mechanisms. For instance, a transfer factor from soil to plant of ¹⁴C would be not be representative for the main mechanism of uptake of ¹⁴C by plants, which is photosynthetic assimilation from the surrounding atmospheric air, together with stable carbon.

During the last years, substantial efforts have been made within the SKB and Posiva research programs in order to develop more appropriate models of ¹⁴C behaviour in the biosphere following underground releases from radioactive waste repositories. The models being developed within these research programmes are dynamic models based on the cycling of stable carbon in different ecosystems. As the development of these models is still ongoing and the resulting models are likely to require a substantial amount of site specific data, which are still being collected, it was considered appropriate to develop a set of simplified, although more conservative models, which could be used in the SFR 1 SAR-08 /SKB 2008/ and KBS-3H /Smith et al. 2007/ assessments.

The objective of the work described in this report was to develop a set of simple conservative models of the environmental behaviour of ¹⁴C released from underground repositories that could be used in the above-mentioned safety assessments. It was the intention that these models would be consistent with approaches commonly applied in assessments of the impact of releases of this radionuclide from other nuclear installations, such as nuclear power plants. The models are, therefore, based on the specific activity approach /Killough 1980/. The developed set of models is applicable for assessments of releases to biosphere receptors and exposure pathways that are of relevance in safety assessments of existing and planned radioactive waste repositories in Finland and Sweden. Figure 1-1 summarises the receptors considered: agricultural lands, forests, lakes, sea basins and irrigation water. In the cases of releases to lakes and sea basins, the models also consider the contamination of sediments, as this may lead to long term human exposure after aggradations of the lakes and sea basins, when sediments may become used for agriculture or forestry. The receptor irrigation water represents the situation when releases result in contamination of fresh waters that are used for irrigation, this could be the case of contamination of wells, lakes and rivers.



Figure 1-1. Receptors of the releases and scenarios considered for the assessment of exposures subsequent to ${}^{14}C$ releases to the biosphere.

In most scenarios considered in safety assessments of radioactive waste repositories, ¹⁴C is released from the waste at low rates over long periods of time. On the other side, the carbon turnover in the environment is relatively fast. Hence, the exposure can be assessed by means of relatively simple equilibrium models. Although very unlikely, in some scenarios it is assumed that ¹⁴C enters the biosphere within a very short time. Under those circumstances, pronounced time-dependence of radionuclides in the environmental compartments could take place. The concentration of a radionuclide in any compartment at equilibrium is numerically identical to the time-integrated concentration after a single pulse-like input into the system, for the same total input. At the same time, the average annual dose over lifetime is the magnitude of interest, which implies an integration period of at least 50 years for adults. But, since the ¹⁴C rate constants in dose-contributing environmental compartments are short compared to the averaging period, even for this case the application of a complex dynamic model is not necessary and equilibrium models can be used. It should be noted, that the turnover times of ¹⁴C in some environmental compartments, like soils and sediments, could be longer. But these compartments usually have an insignificant direct contribution to the doses.

The exposure pathways considered in the dose calculations were: ingestion of contaminated food and water for both terrestrial and aquatic ecosystems, inhalation of contaminated air for terrestrial ecosystems. The exposure by external irradiation has been neglected, as ¹⁴C is a pure low energy beta emitter.

The report provides an overview of the behaviour of ¹⁴C in the environment (Chapter 2) including an outline of conceptual assumptions that are common for all models. This is followed by a detailed description of the terrestrial (Chapter 3) and aquatic (Chapter 4) models. The description of the models includes: 1) specific assumptions, 2) mathematical equations, including those used for dose calculations, 3) example values for all model parameters, including nominal values to be used in deterministic assessments and probability density functions to be used in probabilistic simulations, 4) results of sensitivity and uncertainty analyses. The report ends with a discussion on the applicability of the models and on possible ways for their improvement.

2 Conceptual model of ¹⁴C behaviour in the environment

The most important volatile carbon compounds are methane and carbon dioxide. Additionally, a large variety of volatile organic compounds can be found. These, however, occur only in trace amounts and are insignificant in terms of mass balance in the system. Methane, being released from a deep geological formation may enter the biosphere and be oxidised to carbon dioxide via a number of interim steps /Boeckx and Cleemput 1996/. The key process for the fate of carbon dioxide in the biosphere is photosynthesis, during which CO₂ is converted to carbohydrates. Photosynthesis depends on the environmental conditions as e.g. time of the day, insolation, temperature, water and nutrients supply and plant species. Part of the carbohydrates is lost during respiration to enable the metabolism of the plant; part of it is stored in specific organs as e.g. tubers or grain /Gisi 1990, Sheppard et al. 2006ab/. Table 2-1 summarises typical values and ranges of the net productivity of dry matter biomass and the biomass inventories of a number of ecosystems in different climates /Strasburger 2002/. The net productivity is defined as the difference between gross productivity and respiration.

The net productivity and total biomass inventories vary widely depending on the environmental conditions. The ratio of the net productivity to the total biomass inventory gives an idea of the turnover rate of organic carbon at a site. The turnover rates vary for both terrestrial and aquatic environments by about a factor of 10.

Ecosystem	Net primary productivity (g m ⁻² y ⁻¹)		Biomass (kg m ⁻²)		Turnover (y ⁻¹)
	Typical value	Range	Typical value	Range	
Tropical rainforest	2,200	1,000–3,500	45	6–80	0.020
Green monsoon forest	1,600	1,000–2,500	35	6–60	0.022
Temperate rainforest	1,300	600–2,500	35	6–200	0.027
Deciduous forest	1,200	600–2,500	30	6–60	0.025
Boreal forest	800	400–2,000	20	6–40	0.025
Savannah	900	200–2,000	4	0.2–15	0.0044
Temperate veld	600	200–1,500	1.6	0.2–5	0.0027
Tundra	140	10–400	0.6	0.1–3	0.0043
Semi desert	90	10–250	0.7	0.1–4	0.0078
Extreme desert	3	0–10	0.02	0–0.2	0.0067
Agricultural land	650	100–3,500	1	0.4–12	0.0015
Bog, marsh	2,000	80–3,500	15	3–50	0.0075
Rivers, lakes	250	100–1,500	0.02	0–0.1	8.0E-05
River mouth	1,500	200–1,500	1	0.01–6	6.7E-04
Continental shelf	360	200–600	0.01	0.001–0.04	2.8E-05
Open ocean	125	2–400	0.003	0.005	2.4E-05

Table 2-1. Dry matter net primary productivity and dry matter biomass inventories of different ecosystem types /Strasburger 2002/.

The main idea behind the models proposed here is that the long term environmental behaviour of ¹⁴C is modulated by the environmental cycles of stable carbon (¹²C). It is assumed (*realistic assumption*) that isotopic equilibrium between ¹⁴C and ¹²C is achieved and that a constant isotopic ratio (specific activity) is established, i.e. the same specific activity will be observed in all system components. The specific activity is defined as the activity concentration of ¹⁴C in a medium, expressed in Bq/g or Bq/m³, divided by the stable carbon content in the same media, expressed in g C/g or g C/m³. Isotopes of an element will generally behave in the same way in different processes. An exception to this rule is when isotopic fractionation takes place, e.g. for elements with low atomic weight, like Hydrogen, due to the difference between isotopes in kinetic rates. However, fractionation has a low importance for ¹⁴C, as this isotope is less than 20% heavier than the lightest stable isotope ¹²C. For example, /Killough 1980/ reports fractionation atom ratios for carbon environmental transfers that range between 0.955 and 1.

Furthermore, it is assumed that for continuous long-term releases, equilibrium in different components of the system will be reached (*realistic assumption*). This is a realistic assumption for constant long-term releases; as the carbon turnover in the environment is relatively fast (see Table 2-1). For the case of acute releases, pronounced time variation of ¹⁴C concentrations in terrestrial compartments could take place. However, the endpoint of interest is the average annual dose received by the exposed individuals during their lifetime, which is proportional to the time-integrated concentration. The time-integrated concentration after a single pulse-like input equals the concentration obtained at equilibrium for constant continuous releases and can therefore also be estimated with an equilibrium model. Note that the rate of photosynthesis and other relevant rate constants are small in comparison to the averaging time period, i.e. the life time of an individual.

Another assumption implicit to all models is that the ¹⁴C specific activity reduces as ¹⁴C migrates away from the release source (*realistic assumption*). This is a direct consequence of the irreversibility of the isotopic dilution, i.e. re-concentration of ¹⁴C will not occur once it has mixed with a certain amount of ¹²C /Sheppard et al. 2006a/. This means, that if specific activities near the contamination source are used for dose calculations, this will result in conservative estimates.

3 Models for terrestrial ecosystems

This chapter describes the models for assessment of doses to man resulting from underground releases of ¹⁴C to forest and agricultural soils and for the case of contamination by irrigation water. The models are a modification of the so-called specific activity model, proposed by /Killough 1980/, which has been applied by /UNSCEAR 2000/ for estimating radiation doses to the world population from atmospheric ¹⁴C releases. The same type of models are commonly applied in estimations of local doses arising from atmospheric releases from nuclear and radioactive installations /IAEA 2001/.

3.1 Conceptual model for terrestrial systems

A schematic representation of the conceptual model proposed for terrestrial ecosystems is presented in Figure 3-1. The ¹⁴C that is released to the soil reaches the atmosphere, where it is fully mixed with stable carbon (¹²C) in CO₂ form, present in a layer, hereafter called the mixing layer, delimited by the release area and the mixing height. From the mixing layer ¹⁴C enters the food chain together with ¹²C via photosynthetic assimilation by plants. The ¹⁴C specific activity that is established in the air of the mixing layer is conserved along all components of the food chain to man. Simplifying assumptions made, for deriving equations for the specific activity, in addition to the ones presented in Chapter 2, are discussed below. It should be noted that specific activities in this context refer to the excess ¹⁴C/¹²C ratio due to releases from the repository and do not take into account the presence of natural ¹⁴C.

Assumptions

- 1. The totality of ¹⁴C released to the soil reaches the mixing layer as CO₂ and can be assimilated by plants, if released during a period when photosynthesis can take place (see below). In reality, ¹⁴C might be released to the soil, and subsequently to the atmosphere, in other forms not directly available for assimilation by plants. Their rate of transformation into CO₂ will depend on multiple factors, such as the chemical form and soil properties like pH, redox potential and soil structure. In a review by /Avila et al. 2006/ transformation rates of CH₄ to CO₂ of 0.01–4.9 mg/m² d are reported for agricultural aerobic soils. This shows that this is a potentially important process that needs to be considered in more realistic dose assessments (*Conservative assumption*).
- 2. For calculation of ¹⁴C specific activities in biota, ¹⁴C releases to the mixing layer occurring in the period when photosynthesis can take place should be used, as releases in other periods may not be assimilated by the vegetation. Photosynthesis can take place during the vegetation period, which is about half a year and during daytime which is about half a day. However, it is difficult to estimate which exact fraction of the releases to the soil will be assimilated via photosynthesis, as this fraction will depend on several interacting factors. For example, there exist accumulation processes that may result in C0₂ being stored at night and used during daytime. Also, releases may not be evenly distributed over the year. For instance, larger release rates may take place in the summer when daytime is longer. In the model proposed here, it is conservatively assumed that 100% of the release occurs in periods when it can be assimilated by photosynthesis (*Conservative assumption*).

- 3. For calculation of the ¹⁴C specific activity the total amount of ¹⁴C that is lost from the mixing layer has to be estimated. Only the ¹⁴C that is lost from the mixing layer with lateral air exchange caused by wind is considered. Losses of ¹⁴CO₂ to higher atmospheric layers are conservatively neglected (*Conservative assumption*).
- 4. The height of the mixing layer is taken as the height needed to supply the canopy with CO₂ demand. A well developed canopy, which is able to assimilate daily 2–3 g CO₂/m² soil in a sunny summer day during photosynthesis /Geisler 1980/, requires the ¹²CO₂ contained in a 20 m height. However, on sunny days, the effective mixing height could be much higher. The insolation causes a convective boundary layer, the height of which increases up to 1,000 m from the morning to the early afternoon /Grünhage et al. 1999, Grünhage and Jäger 2001/. Also, there is a gradient of CO₂ in the air, since, due to photosynthesis, the canopy is an effective CO₂-sink that causes a permanent flux of CO₂ from upper atmosphere layers to the ground. These effects are not taken into account in this model (*Conservative assumption*).
- 5. The assimilation of carbon by plants via root uptake can be neglected as it is only 10 percent or less the total carbon assimilation, which is mainly via photosynthesis see /Avila et al. 2006/ (*Realistic assumption*).



Figure 3-1. Schematic representation of the conceptual model proposed for assessment of exposures to ¹⁴C released from the geosphere to terrestrial ecosystems (forests and agricultural lands). The arrows represent fluxes of ¹⁴CO₂ and ¹²CO₂. Fluxes between the mixing layer (see assumption 4 in this chapter) and higher atmospheric layers are conservatively neglected. The model assumes that the same ${}^{14}CO_2/{}^{12}CO_2$ ratio is established in the air and the canopy.

3.2 Mathematical model

Under the above assumptions, for the case of continuous ¹⁴C releases to the soil the excess ¹⁴C/¹²C ratio (specific activity) at equilibrium can be calculated with the following equation:

$$R_{{}^{14}C/{}^{12}C} = \frac{E \cdot \frac{\text{Re}\,\text{leaseRate}}{Area}}{h \cdot \lambda_{ex} \cdot C_{{}^{12}C} + NPP}$$
(3.1)

where,

 $R_{14C/12C}$ is the excess ¹⁴C/¹²C ratio – specific activity (Bq/g),

E is the effective release fraction – fraction of ¹⁴C release to the mixing layer that occurs in a period when photosynthesis can take place (unit less),

ReleaseRate is the release rate of ¹⁴C to soil from the geosphere (Bq/y),

Area is the release area (m^2) ,

h is the mixing height (m),

 λ_{ex} is the air exchange rate in the mixing layer (1/y),

 C_{12C} is the ¹²C content in air (g C/m³),

NPP is the net primary production in the ecosystem (g $C/m^2/y$).

The rate constants of the processes involved in photosynthesis are very short compared to the averaging period (see Chapter 2). Hence, over a given time period (T_{av}) the average excess ¹⁴C/¹²C-ratio for a single pulse-like release can be calculated according to:

$$R_{{}^{14}C/{}^{12}C} = \frac{E \cdot \frac{\text{Re}\,\text{lease}}{Area}}{(h \cdot \lambda_{ex} \cdot C_{{}^{12}C} + NPP) \cdot T_{av}}$$
(3.2)

where,

 $R_{14C/12C}$ is the excess ¹⁴C/¹²C ratio – specific activity (Bq/g),

E is the effective release fraction – fraction of the ¹⁴C release to the mixing layer that occurs in the period when photosynthesis can take place (unit less),

Release is the release of ¹⁴C from the geosphere to the soil (Bq),

Area is the release area (m^2) ,

h is the mixing height (m),

 λ_{ex} is the air exchange rate in the mixing layer (1/y),

 C_{12C} is the ¹²C content in air (g C/m³),

 T_{av} is the averaging period (y),

NPP is the net primary production in the ecosystem (g $C/m^2/y$).

The air exchange rate in the mixing layer can be obtained by dividing the wind speed at the vegetation height by the ratio of the affected area:

$$\lambda_{ex} = \frac{v}{r} \tag{3.3}$$

where,

v is the wind speed at the vegetation height (m/y),

r is the ratio of the release area (m).

Assuming that the release area has a circular form, it's ratio can be calculated with the following equation:

$$r = \sqrt{Area/\pi} \tag{3}$$

where,

r is the ratio of the release area (m),

Area is the release area (m^2) .

In Equation 3.3 the wind speed at the vegetation height is used. The vegetation height is defined as the height at which the photosynthetic assimilation of carbon is most efficient. Empirical values of the wind speed at this height are usually not available, but can be estimated from commonly measured values at the height of 10 m (v_{10}), assuming an exponential wind profile and a vegetation-specific roughness length z_0 /Seinfeld 1986/. The roughness length is defined as the height at which the wind speed becomes zero when the wind profile above the canopy is extrapolated:

$$v = v_{10} \cdot \frac{\ln(\frac{h_{veg}}{z_0})}{\ln(\frac{10}{z_0})}$$
(3.5)

where,

v is the wind speed at the vegetation height (m/y),

 v_{10} is the wind speed at the height of 10 m (m/y),

 h_{veg} is the vegetation height (m),

 z_0 is the roughness length (m).

Application to irrigation

In the case of irrigation of agricultural lands with water contaminated with ¹⁴C, plants may also incorporate ¹⁴C from the air via photosynthesis.

The incorporation via photosynthesis can be estimated by calculating the excess ${}^{14}C/{}^{12}C$ ratio (specific activity) at equilibrium using Equation 3.1, where the release rate (*ReleaseRate*) is calculated with Equation 3.6. This equation assumes that all ${}^{14}C$ that is inputted to the system with irrigation water will be immediately released to the mixing layer where it can be assimilated by the irrigated plants via photosynthesis.

 $ReleaseRate = Conc_{irrWater} \cdot Volume_{irr} \cdot Area$

where,

ReleaseRate is the input of ¹⁴C with contaminated irrigation water (Bq/y),

Conc_{irrWater} is the ¹⁴C concentration in irrigation water (Bq/m³),

*Volume*_{irr} is the total volume of irrigation water per unit area used in a year (m/y),

Area is the irrigated area (m²).

Note that the parameter *Area* in Equation 3.6 has the same meaning as the release area in Equation 3.1. Hence, this parameter cancels off when the *ReleaseRate* in Equation 3.1 is replaced with Equation 3.6 and the following equation is obtained for the ${}^{14}C/{}^{12}C$ ratio (specific activity):

$$R_{{}^{14}C/{}^{12}C} = \frac{Conc_{irrWater} \cdot Volume_{irr}}{h \cdot \lambda_{ex} \cdot C_{{}^{12}C} + NPP}$$
(3.7)

(3.6)

(3.4)

where,

 $R_{14_C/12_C}$ is the excess ¹⁴C/¹²C ratio – specific activity (Bq/g),

Conc_{irrWater} is the ¹⁴C concentration in irrigation water (Bq/m³),

Volume_{irr} is the total volume of irrigation water per unit area used in a year (m/y),

h is the mixing height (m),

 λ_{ex} is the air exchange rate in the mixing layer (1/y),

 C_{12_C} is the ¹²C content in air (g C/m³),

NPP is the net primary production in the ecosystem (g $C/m^2/y$).

The irrigation water could be water from a well, a lake or a river that is contaminated with ¹⁴C. The ¹⁴C concentration in the lake (river) water can be calculated with the equations given in Chapter 4. The concentration in well water can be estimated by dividing the release rate by the well capacity.

Dose calculations

For releases to forest and agricultural lands (including inputs via irrigation) annual doses via inhalation and food ingestion are calculated using the specific activities obtained with the above Equations (3.1, 3.2 and 3.7) as follows:

$$Dose_{ingFood} = R_{14_{C}/12_{C}} \cdot IR_{C} \cdot DCC_{ingFood}$$
(3.8)

where,

Dose_{ingFood} is the dose from intake of ¹⁴C with ingested food (Sv/y),

 $R_{14_C/12_C}$ is the excess ¹⁴C/¹²C ratio – specific activity (Bq/g C),

 IR_C is the intake rate of carbon by adults (g C/y),

DCC_{ingFood} is the dose conversion coefficient for calculation of doses via food ingestion (Sv/Bq).

The number of people that could be exposed to ¹⁴C via food ingestion, at the levels obtained with Equation 3.8, can be estimated as:

$$N = \frac{pty \cdot Area}{IR_c}$$
(3.9)

where,

N is the number of people exposed via food ingestion (unitless),

pty is the productivity of edible food at the contaminated object (g C/m²/y),

Area is the area of the contaminated object – release area (m²),

 IR_C is the annual carbon intake by people (g C/y).

$$Dose_{inh} = R_{l_{4C/l_{2C}}} \cdot C_{l_{2C}} \cdot InhR \cdot ExposTime \cdot DCC_{inh}$$
(3.10)

where,

 $Dose_{inh}$ is the dose from incorporation of ¹⁴C via inhalation (Sv/y),

 $R_{14C/12C}$ is the excess ¹⁴C/¹²C ratio-specific activity (Bq/g C),

 C_{12C} is the ¹²C content in air (g C/m³),

InhR is the inhalation rate (m^3/h) ,

ExposTime is the fraction of the time that the individual is exposed via inhalation to the contamination (h/y),

 DCC_{inh} is the dose conversion coefficient for calculation of doses via inhalation (Sv/Bq).

3.3 Parameter values

For each of the model parameters, a nominal value – to be used in deterministic simulations and a probability density function (PDF) – to be used in probabilistic simulations, are given in Table 3-1. For some parameters only conservative values are given and PDFs are not assigned, as the conservative values were chosen to guarantee that there is very low probability that a realistic value would be above (for parameters that have a positive correlation to the doses) or below (for parameters with negative correlation to the doses) this value. As far as possible, parameter values have been chosen to be representative for (candidate) repository sites in Finland and Sweden.

Values of the Dose Conversion Coefficients for food ingestion ($DCC_{ingFood}$) and inhalation (DCC_{inh}), used for dose calculations with Equations 3.8 and 3.10, were taken from /Avila and Bergström 2006/, which are values recommended in /ICRP 1996/:

 $DCC_{ingFood} = 5.8.10^{-10} \text{ Sv/Bq}$ $DCC_{inh} = 5.8.10^{-9} \text{ Sv/Bq}$

The appropriate value for the averaging period (T_{av}) will depend on the period over which the annual doses are averaged. For instance, if the average lifetime annual doses for an adult person are to be estimated then an averaging period of 50 y is appropriated, which is the integration period used in /ICRP 1996/ for derivation of the DCC for adults. For estimation of average lifetime doses to children the averaging period should be longer. In general, the averaging period should correspond to the integration period used in the derivation of the DCC used in the dose calculation.

Comments on the parameter values

For some of the model parameters additional comments are given below regarding how the values presented in Table 3-1 were selected.

Wind speed and height of the mixing layer

The wind speed and the height of the mixing layer both depend on the local meteorological conditions at the site, which are affected by the type of vegetation canopy. As mentioned in the description of the conceptual model (assumption 4), the mixing height is taken as the height needed to supply the canopy with the daily intake of carbon. The daily intake of carbon by the canopy experiences variation between days and sites, which would result in variation in the value given to the mixing height. These variations are taken into account by assigning a probability distribution to the mixing height. However, it should be taken into account that the "real" mixing height is probably much higher than the conservative values assumed in this study. During calms and periods with very low wind speeds there is a preferential vertical ${}^{12}CO_2$ flux that supplies the canopy. In periods with higher wind speeds, the ${}^{12}CO_2$ flux to the canopy is dominated by a horizontal exchange due to the action of the wind. This interaction between the height of the mixing layer and the wind speed can be taken into account in probabilistic simulations by introducing a negative correlation between these parameters. To assess the potential effect of such interaction we performed simulations assuming different correlation coefficients, varying from -1 to 0. There was a marginal difference, of less than 10%, in the results of these simulations, which indicates that the effect of this interaction can be neglected in the context of this model.

The wind speed is used in the model for calculating the exchange rate of the air in the mixing layer, which is needed for estimating the ¹⁴C specific activity. The wind speed is commonly measured at a standard height of 10 m. But in the model the wind speed at the vegetation height is used. The vegetation height is defined as the height at which the photosynthesis is most efficient, which is assumed to be equal to the canopy height, where light interception is

Table 3-1. Examples of nominal values, for use in deterministic simulations, and probability density functions (PDF), for use in probabilistic simulations, for the parameters of the ¹⁴C model for terrestrial ecosystems. The model equations where these parameters are included are indicated. The parameter values are reported in units in which these are commonly measured and reported in the literature.

Parameter	Units	Equations	Nominal value	PDF(*)	Comments
Area Release area	m²	3.1, 3.2, 3.4, 3.6, 3.9			Situation specific value required
E Effective release fraction	unit less	3.1, 3.2	0.5		Conservative value (see assumption 2 in Section 3.1)
V_{10} Wind speed at 10 m	m/s	3.5	5	Weibull (5, 1.8)	Typical value at 10 m for Northern Europe /Troen and Lundtang Petersen 1989/
z _o Roughness length for agricultural lands	m	3.5	0.25		Value given in /Seinfeld 1986/ and /Mayall 2003/ for farmlands
z_0 Roughness length for forests	m	3.5	1		Value given in /Seinfeld 1986/ and /Mayall 2003/ for forests
<i>h</i> _{veg} Vegetation height for agricultural lands	m	3.5	1		Value given in /Seinfeld 1986/ and /Mayall 2003/ for farmlands
h_{veg} Vegetation height for forests	m	3.5	2		Value given in /Seinfeld 1986/ and /Mayall 2003/ for forests
h	m	3.1, 3.2	10	LGN (10, 2)	Value for agricultural lands
Mixing height for agricultural lands				Truncated at 1 m	See assumption 4 in Section 3.1
h	m	3.1, 3.2	20	LGN (10, 2)	Value for forests
Mixing height for forests				Truncated at 2 m	See assumption 4 in Section 3.1
<i>IR_c</i> Carbon intake rate	g C/y	3.8, 3.9	1.1E+5		/ICRP 1975, Avila and Bergström 2006/
<i>Volume_{irr}</i> Irrigation volume	m/y	3.6, 3.7	0.15	Triang (0.1, 0.2, 0.15)	Value given in /Bergström and Barkefors 2004/
<i>NPP</i> Net primary production of the vegetation forests	g C/m²/y	3.1, 3.2	250	N (250, 25)	Value given in /Lindborg (ed) 2005/ for forest in the Forsmark area
NPP Net primary production of the vegetation Agricultural lands	g C/m²/y	3.1, 3.2	120	N (120, 12)	Value given in /Lindborg (ed) 2005/ for agricultural lands in the Forsmark area
NPP Net primary production of the vegetation Irrigation	g C/m²/y	3.7	98	Triang (73.5, 196, 98)	Obtained by multiply- ing values of yields for vegetables (in g FW/m²/y) given in /Bergström and Barkefors 2004/ by the carbon content of vegetables (0.049 g C/g FW)
<i>ExposTime</i> Fraction of time exposed via inhalation	h/y	3.10	2,920		Assuming 8 hours every day, which could be reasonable for an agricultural worker
<i>inhR</i> Inhalation rate	m³/h	3.10	1		/ICRP 1975, Avila and Bergström 2006/

(*) For the weibull distribution the first parameter is the Mean and the second the Shape Factor. For the lognormal (LGN) and normal (N) distributions the first parameter is the Mean and the second the Standard Deviation of the untransformed data. For the triangular distribution (Triang) the first parameter is the Minimum, the second the Maximum and the third the Mode.

most intensive. As the canopy height varies from case to case, this will result in variations in the vegetation height. The assumed exponential dependency of the wind speed with height is well established /Seinfeld 1986/. However, parameters needed for the extrapolations with Equation 3.5 (the vegetation height and the roughness length) can vary by a factor of 100 between different types of surfaces. This is illustrated in Table 3-2 where values reported in /Seinfeld 1986/ and /Mayall 2003/ for different surface types are presented. The value of the vegetation height given in these publications for forests seems to be low. However, a higher value would result in higher wind speeds and therefore in lower ¹⁴C specific activities. It should be noted that average wind speeds in the mixing layer, estimated using the values giving in Table 3-1 for v_{10} , z_0 and h, are about two times lower than the values of the wind speed at the vegetation height. The average wind speeds in the mixing layer were obtained by integrating Equation 3.5 from the roughness height to the height of the mixing layer and dividing the result by the mixing height.

3.4 Sensitivity and uncertainty analyses

A sensitivity analysis was carried out to study the effect on the model predictions of variations in the parameters. At the same time, an uncertainty analysis was conducted to propagate parameter uncertainties throughout the models – to obtain an estimate of the overall parameter uncertainty of the model predictions. For this purpose, probabilistic simulations (10,000 samples) using Latin Hypercube sampling were carried out with the software package Eikos /Ekström and Broed 2006/. The Spearman Rank Correlation Coefficients (SRCC) were computed as sensitivity indexes. The SRCC is an appropriate sensitivity measure for models with monotonic relationships between parameters and outputs, which is the case for the studied models. Three cases were considered in the sensitivity study where for parameters that are usually easy to obtain for each specific situation, such as the area of the agricultural land and forests and the capacity of the well used for irrigation, the values were taken from the SFR 1 SAR-08 /SKB 2008/ safety assessment.

The three cases considered were:

- 1. Continuous unit release rate (1 Bq/y) to a forest ecosystem with an area of $1.37\text{E6} \text{ m}^2$.
- 2. Continuous unit release rate (1 Bq/y) to an agricultural with an area of 1.37E6 m².
- 3. Continuous unit release rate (1 Bq/y) to a well that is used for irrigation on agricultural land, with an area of 1E4 m², which is used for production of vegetables. The value of the irrigated area was taken as the area that can be irrigated with water from a well of a given capacity. The concentration of ¹⁴C in the irrigation water was calculated by dividing the release rate by the well capacity. Values of well capacities (l/h) reported for the Forsmark area were fitted to a lognormal normal distribution (with the following percentiles: 5% 35 l/h, 50% 468 l/h, 95% 6,188 l/h) and used in the probabilistic simulations with a lower constrain of 57 l/h.

Table 3-2.	Roughness	length and	vegetation	height for	various	vegetation of	anopies
/Seinfeld 1	986, Mayall	2003/.	_	-		-	

Surface	Roughness length (m)	Vegetation height (m)
Lawn	0.01	0.05
Uncut grass	0.05	0.2
Fully grown root crops	0.1	0.4
Farmland	0.25	1
Forest	1	2

For other parameters the PDF, or the nominal value, when a PDF was missing (see Table 3-1) were used in the simulations. For the irrigation case an intake rate of carbon (IR_c) of 2,080 g C/y was used, which is the value that was used in the SFR 1 SAR-08 /SKB 2008/ safety assessment.

The statistics of the total annual doses per unit release rate (doses from ingestion of food and inhalation) obtained for the three studied cases are presented in Table 3-3. The values obtained for the agricultural land and the forest are close to each other and in both cases the ratio between the 95 and 5 percentiles is around 10. In the case of irrigation a larger interval of variation of around 200 was observed, which can be explained by the large assumed variability of the well capacity.

The contributions of ingestion of food and inhalation to the total annual doses per unit release rate obtained for the three studied cases are shown in Table 3-4. The inhalation doses for the forest and the agricultural land were very similar as the specific activities obtained for these two cases are very close to each other (see below). For the three cases the food ingestion doses are one order of magnitude or more higher than the values from inhalation, which is consistent with values reported in other studies /UNSCEAR 2000/.

The statistics of the ${}^{14}C/{}^{12}C$ ratios (specific activity) obtained for the three studied cases are presented in Table 3-5. The relationship between the specific activities obtained for the three studied cases is the same as the relationship between the annual doses (see Table 3-3).

Table 3-3. Statistics of the total annual doses (in Sv/y) obtained for the three calculation cases: continuous releases of 1 Bq/y to a forest, an agricultural land and a well used for irrigation.

Case	Mean	SD	5%	50%	95%
Forest	7.8E–16	1.3E–15	2.0E-16	4.9E-16	2.1E–15
Agricultural land	6.3E-16	1.2E–15	1.6E–16	3.9E-16	1.7E–15
Irrigation	1.1E–16	4.5E-16	2.2E-18	3.6E-17	4.2E-16

Table 3-4. Mean values of the annual doses from food ingestion and inhalation (in Sv/y) obtained for the three calculation cases: continuous releases of 1 Bq/y to a forest, an agricultural land and a well used for irrigation.

Case	Food ingestion	Inhalation	
Forest	7.4E–16	3.5E–17	
Agricultural land	6.0E-16	2.8E-17	
Irrigation	1.1E–16	9.0E–18	

Table 3-5. Statistics of the ¹⁴C¹²C Ratios (specific activity in Bq/g C) obtained for the three calculation cases: continuous releases of 1 Bq/y to a forest, an agricultural land and a well used for irrigation.

Case	Mean	SD	5%	50%	95%
Forest	1.2E–11	2.3E-11	3.0E-12	7.3E–12	3.2E-11
Agricultural land	9.4E-12	1.7E–11	2.4E-12	5.8E-12	2.6E-11
Irrigation	8.8E-11	3.4E-10	1.7E–12	2.8E-11	3.2E-10

The results of the sensitivity study are shown in Figures 3-2 and 3-3. In the cases of releases to agricultural lands and forests, the parameter that has the highest influence on the variation of the predicted annual doses is the wind speed, which defines the lateral removal of radionuclides from the mixing layer. This parameter is relatively easy to measure and there is usually site specific data available. In Figure 3-4 results of measurements of the wind speed at the height of 20 m obtained in Olkiluoto, Finland are presented as a histogram. A good fit to a Weibull distribution was obtained (curve in Figure 3-4) with parameter values (Mean = 4.21 and Shape Factor = 24) that are very close to the values recommended in Table 3-1. Other parameters have a substantially lower influence on the dose predictions. For both, agricultural land and forest, the height of the mixing layer has also some influence, but this parameter has been given conservative values and thus obtaining site specific values is not as important as for the wind speed.

For the case of irrigation (Figure 3-3), the well capacity has a prominent effect on the predicted specific activities, and doses. However, the uncertainty in the well capacity is not relevant for the models studied here. It can be seen from this figure that the wind speed is the model parameter with the highest effect on the specific activities, as for the cases of direct releases to forests and agricultural lands.



Figure 3-2. Spearman Rank Correlation Coefficients (SRCC) of the Total ¹⁴C annual doses per unit release rate obtained for the cases of releases to a forest (left figure) and an agricultural land (right figure). The parameter names can be found in Table 3-1.



Figure 3-3. Spearman Rank Correlation Coefficients (SRCC) of the ${}^{14}C/{}^{12}C$ ratios (specific activities) obtained for the case of irrigation. The parameter names can be found in Table 3-1.



Figure 3-4. Histogram of values of the wind speed at 20 m height measured in the Olkiluoto, Finland and curve obtained from fitting the measured values to a Weibull distribution. The vertical lines indicate the 5 and 95 percentiles of the fitted distribution.

4 Models for aquatic ecosystems

This chapter describes the models for assessment of doses to man resulting from underground releases of ¹⁴C to lakes and sea basins. As mentioned in Chapter 3, in assessments of the radio-logical impact of ¹⁴C releases to terrestrial ecosystems, models based on the specific activity approach, proposed by /Killough 1980/, are commonly applied. A modification of this approach was used in the models proposed in Chapter 3 for assessment of underground releases directed to agricultural lands, forests and for indirect releases to agricultural lands via irrigation. The same conceptual approach (see Chapter 2) is applied in this chapter for the case of underground releases to aquatic ecosystems, covering also situations where aquatic ecosystems are transformed into terrestrial ecosystems, for example as a result of aggradations of lakes, land rise, etc.

4.1 Conceptual model for aquatic ecosystems

A schematic representation of the conceptual model proposed for aquatic ecosystems is presented in Figure 4-1. The ¹⁴C released to the bottom sediment of a lake or sea basin reaches the water layer, where it is fully mixed with stable inorganic carbon (¹²C) dissolved in water (DIC). The ¹⁴C dissolved in water enters the food chain together with ¹²C (DIC) via photosynthetic assimilation by primary producers, mainly phytoplankton and benthic plants. The same value of the ¹⁴C specific activity established in the water layer is conserved along all components of the food chain to man. The simplifying assumptions, in addition to the ones presented in Chapter 2, made for deriving equations for calculating specific activities are discussed below. It should be noted, that the specific activities in this context refer to the excess ¹⁴C/¹²C ratio due to the releases from the repository and do not take into account the presence of natural ¹⁴C.

Assumptions

1. It is assumed that, for releases occurring when photosynthesis is active (see below), the totality of the 14 C released to the sediment reaches the water layer as CO₂ or in other forms that can be readily assimilated by primary producers. In reality, ¹⁴C might be released in other forms that are not directly available for assimilation by plants, for example as methane (CH₄). Methane oxidation in lakes has been reported in a number of studies /Venkiteswaran and Schiff 2005, Casper et al. 2003, Heilmann and Carlton 2001/. The dynamics of methane production and oxidation in the sediments of the Kiel Harbour were studied over a period of 2 years by /Schmaljohann 1996/. Data of this study do not allow the determination of what proportion of the methane produced is actually oxidized at the sediment surface, but show that aerobic methane oxidation is an important oxygen-consuming process in this environment. The conversion rates of methane depend on the redox conditions in the water column. Seasonal variations are reported by /Utsumi et al. 1998/ in a eutrophic lake with low conversion rates from January to April and high conversion rates in autumn. In total, 74% of the methane dissolved in water in this lake is converted to CO₂. Relevant methane oxidation and methane evasion has been also reported by /Rudd and Hamilton 1978/, who observed pronounced seasonality of methane oxidation in a eutrophic Canadian Shield lake. Methane oxidation peaked in the fall and was very low during the rest of the year. Methane and carbon dioxide releases from a small lake have been studied by /Casper et al. 2000/. The study showed that methane was released mainly by ebullition, whereas carbon dioxide was lost by diffusion. Seasonal dependencies of the methane oxidation rates in the sea environment have also been reported /Schmaljohann 1996/ (Conservative assumption).

- 2. For the calculation of the ¹⁴C specific activity in biota, ¹⁴C releases to the water layer in the period when photosynthesis can take place should be used, as releases in other periods will not be assimilated by primary producers. The photosynthesis rates are not homogeneous over the year. For instance, in winter photosynthesis rates are usually lower. Moreover, as photosynthesis occurs only above a certain illumination, there is no photosynthesis during the night. The processes influencing the photosynthesis rates are complex and difficult to quantify. As for the terrestrial model, it is conservatively assumed that 100% of the release to the water layer occurs in periods when it can be assimilated by photosynthesis (*Conservative assumption*).
- 3. For the calculation of the ¹⁴C specific activity, the amount of ¹⁴C that is transported out from the water layer has to be estimated. Only the ¹⁴C that is transported with water fluxes is considered. Losses of ¹⁴C to the atmosphere are conservatively neglected (*Conservative assumption*).



Figure 4-1. Schematic representation of the conceptual model proposed for assessment of exposures to ¹⁴C released from the geosphere to aquatic ecosystems (lakes and sea basins). The arrows represent fluxes of ¹⁴CO₂ and ¹²CO₂. Fluxes between the water layer and the atmosphere are conservatively neglected. The model assumes that the same ¹⁴CO₂/¹²CO₂ ratio in the water and the primary producers established.

4.2 Mathematical models

Under the above assumptions, for the case of continuous ¹⁴C releases to lakes the excess ¹⁴C/¹²C ratio (specific activity) at equilibrium can be calculated with the following equation:

$$R_{{}^{14}C/{}^{12}C} = \frac{E \cdot \frac{\text{ReleaseRate}}{Area}}{DIC \cdot \frac{AreaCatch}{Area} \cdot runoff + NPP}$$
(4.1)

where,

 $R_{14C/12C}$ is the excess ¹⁴C/¹²C ratio (Bq/g C),

E is the effective release fraction – fraction of the 14 C release that occurs in the period when photosynthesis can take place (unit less),

ReleaseRate is the release rate of ¹⁴C from the geosphere to the lake (Bq/y),

Area is the lake area (m²),

DIC is the concentration of dissolved inorganic carbon (DIC) in the lake water ($g C/m^3$),

AreaCatch is the catchment area of the lake (m²),

runoff is the runoff in the catchment area of the lake (m/y),

NPP is the lake net primary production (g $C/m^2/y$).

In the case of continuous releases to sea basins the excess ${}^{14}C/{}^{12}C$ ratio (specific activity) at equilibrium can be calculated with a similar equation:

$$R_{{}^{14}C/{}^{12}C} = \frac{E \cdot \frac{\text{Re}\,\text{leaseRate}}{Area}}{DIC \cdot \frac{Depth}{'RT} + NPP}$$
(4.2)

where,

 $R_{I_{4C}/I_{2C}}$ is the excess ¹⁴C/¹²C-Ratio (Bq/g C),

E is the effective release fraction – fraction of the 14 C release that occurs in the period when photosynthesis can take place (unit less),

ReleaseRate is the release rate of ¹⁴C from the geosphere to the sea basin (Bq/y),

Area is the area of the sea basin (m^2) ,

DIC is the concentration of dissolved inorganic carbon (DIC) in the sea water (g C/m³),

Depth is the mean depth of the sea basin (m),

RT is the residence time of water in the sea basin (y),

NPP is the net primary production in the sea basin (g $C/m^2/y$).

The relationship between the excess ¹⁴C/¹²C-Ratio and the volume and depth of the lake and sea basin is not obvious from Equations 4.1 and 4.2. However, these equations can be reformulated to illustrate the relationship with these properties by substituting the area of the lake and the sea basin by the ratio between the volume and the mean depth of the lake and the sea basin respectively. The same is valid for Equations 4.3 and 4.4 below.

The characteristic times of the kinetic processes involved in photosynthesis are very short compared to the averaging period (see Chapter 2). Hence, over a given time period the average excess $^{14}C/^{12}C$ -ratio for single pulse-like releases can be calculated with Equation 4.3 for lakes and with Equation 4.4 for sea basins:

$$R_{{}^{14}C/{}^{12}C} = \frac{E \cdot \frac{\text{Release}}{Area}}{(DIC \cdot \frac{AreaCatch}{Area} \cdot runoff + NPP) \cdot T_{av}}$$

$$R_{{}^{14}C/{}^{12}C} = \frac{E \cdot \frac{\text{Release}}{Area}}{(DIC \cdot \frac{Depth}{RT} + NPP) \cdot T_{av}}$$

$$(4.3)$$

where,

 $R_{14_C/12_C}$ is the excess ¹⁴C/¹²C-Ratio (Bq/g C),

E is the effective release fraction – fraction of the ¹⁴C release that occurs in the period when photosynthesis can take place (unit less),

Release is the release of ¹⁴C from the geosphere to the lake or sea basin (Bq),

Area is the lake or sea basin area (m²),

DIC is the concentration of dissolved inorganic carbon in the lake or sea water (g C/m³),

AreaCatch is the catchment area of the lake (m²),

runoff is the runoff in the catchment area of the lake (m/y)

NPP is the lake net primary production in the lake or the sea basin (g $C/m^2/y$),

Depth is the mean depth of the sea basin (m),

RT is the residence time of water in the sea basin (y),

 T_{av} is the averaging period (y).

Aggradations of lakes and sea basins

As a result of long-term releases to lakes and sea basins, ¹⁴C may become accumulated in the sediments. Due to land rise, filling of lakes and sea basins, etc terrestrial ecosystems might be established on contaminated lake or sea sediments, either by natural succession processes or as result of future human actions. As the organic matter of the newly formed soils decomposes, the ¹⁴C accumulated in the sediment (soil) will be released to the atmosphere and enter the terrestrial food chains (see Chapter 3). Such situations can be assessed by combining the models described above in this chapter with the models described in Chapter 3. Firstly, the ¹⁴C inventory in sediments has to be estimated. This can be done by multiplying the excess ratios calculated with Equation 4.1 or 4.2 by the carbon accumulation rate in the lake or the sea basin and by the formed terrestrial ecosystem is estimated by multiplying the ¹⁴C inventory in sediment (soil) by the rate of decomposition of the soil organic matter. The following equation is then obtained for the release rate of ¹⁴C in the terrestrial object:

$$ReleaseRate = R_{I4_{C}/I2_{C}} \cdot Sed \cdot T_{release} \cdot Decomp$$
(4.5)

where,

ReleaseRate is the release rate of ¹⁴C from the newly formed terrestrial object (Bq/y),

 $R_{I4_C/I2_C}$ is the excess ¹⁴C/¹²C-Ratio obtained with Equations 4.1 or 4.2 (Bq/g C),

Sed is the carbon accumulation rate in the sediment of the lake or sea basin (g $C/m^2/y$),

Area is the lake or sea basin area (m²),

 $T_{release}$ is the duration of the release to the lake or sea basin (y),

Decomp is the decomposition rate of organic matter in the newly formed terrestrial object (1/y).

The release rate estimated with Equation 4.5 is then used as input in Equation 3.1 to obtain the specific activity in the formed terrestrial ecosystem.

Dose calculations

For releases to lakes and sea basins doses via food ingestion are calculated using the specific activities obtained with the above Equations (4.1, 4.2, 4.3 and 4.4) as follows:

$$Dose_{ingFood} = R_{14_C/12_C} \cdot IR_C \cdot DCC_{ingFood}$$

$$\tag{4.6}$$

where,

Dose_{ingFood} is the dose from intake of ¹⁴C with ingested food (Sv/y),

 $R_{14_C/12_C}$ is the excess ¹⁴C/¹²C ratio (Bq/g C),

 IR_C is the intake rate of carbon by adults (g C/y),

 $DCC_{ingFood}$ is the dose conversion coefficient for calculation of doses received via food ingestion (Sv/Bq).

The number of people that could be exposed to ¹⁴C via food ingestion, at the levels obtained with Equation 4.6, can be estimated as:

$$N = \frac{pty \cdot Area}{IR_c}$$
(4.7)

where,

N is the number of people exposed via food ingestion (unit less),

pty is the productivity of edible food at the contaminated object (g $C/m^2/y$),

Area is the area of the contaminated object (m²),

 IR_C is the annual carbon intake by adults (g C/y).

For the case of releases to lakes, exposures via ingestion of water could also be relevant. It can be expected that in drinking water ¹⁴C will be present only in dissolved inorganic form. The ¹⁴C content in drinking water can then be obtained by multiplying the DIC content by the specific activity. Hence, the doses via water ingestion can be calculated as:

$$Dose_{ingWater} = R_{14_C/12_C} \cdot DIC \cdot IR_{Water} \cdot DCC_{ingWater}$$
(4.8)

where,

 $R_{14_C/12_C}$ is the excess ¹⁴C/¹²C ratio (Bq/g C),

DIC is the concentration of dissolved inorganic carbon in the lake water (g C/m³)

 IR_{Water} is the intake rate of water (m³/y),

 $DCC_{ingWater}$ is the dose conversion coefficient for calculation of doses received via water ingestion (Sv/Bq).

4.3 Parameter values

For each of the model parameters a nominal value, to be used in deterministic simulations, and a probability density function (PDF), to be used in probabilistic simulations, are given in Table 4-1. For some parameters, only conservative values are given and PDFs are not assigned, as the conservative values were chosen to ensure that there is very low probability that a realistic value would be above (for parameters that have a positive correlation to the doses) or below (for parameters with negative correlation to the doses) this value. As far as possible, the parameter values have been chosen to be representative for (candidate) repository sites in Finland and Sweden.

Table 4-1. Examples of nominal values, for use in deterministic simulations, and probability density functions (PDF), for use in probabilistic simulations, for the parameters of the ¹⁴C model for aquatic ecosystems. The model equations where these parameters are included are indicated. The parameter values are reported in units in which these are commonly measured and reported in the literature.

Parameter	Units	Equations	Nominal value	PDF(*)	Comments
<i>Area</i> Lake or sea basin area	m²	4.1, 4.2, 4.3, 4.4, 4.5, 4.7			Situation specific value required
AreaCatch catchment area of the lake	m ²	4.1, 4.3			Situation specific value required
<i>E</i> Effective release fraction	unitless	4.1, 4.2, 4.8	1		Conservative value (assumption 2)
<i>DIC</i> lake water dissolved inorganic carbon	g C/m³	4.1, 4.3, 4.7	22	N (22, 10)	Value given in /Lindborg (ed) 2005/
<i>DIC</i> sea water dissolved inorganic carbon	g C/m ³	4.2, 4.4,	11	N (11, 5)	Value given in /Lindborg (ed) 2005/
Runoff In Lake catchment	m/y	4.1, 4.3	0.226	Triang (0.2, 0.3, 0.226)	Value given in /SKB 2006/
NPP lake Net primary production	g C/m²/y	4.1, 4.3	185	N (185, 18.5)	Value given in /Lindborg (ed) 2005/
NPP sea basins Net primary production	g C/m²/y	4.2, 4.4	122	N (122, 12.2)	Value given in /Lindborg (ed) 2005/
<i>Depth</i> Mean depth of sea basin	m	4.2, 4.4			Situation specific value required
<i>RT</i> residence time in sea basin	У	4.2, 4.4	2.0E-3	Triang (1.0E–3, 4.2E–3, 2.0E–3)	Value given in /Engqvist and Andrejev 1999/
Sed accumulation rate in lake sediments	g C/m²/y	4.5	17	LGN (17, 4)	Estimated from the accumulation rate of 1E–3 m/y /SKB 2003/, measured values of carbon content in sediments /SKB 2004a/ and measured values of water content in sediments /SKB 2007/
<i>T_{release}</i> release duration	У	4.5			Situation specific value required
Decomp Decomposition rate	1/y	4.5	0.03	N (0.03, 0.01)	/Gisi 1990/
<i>IR_c</i> Carbon intake rate	g C/y	4.6, 4.7	1.1E+5		/ICRP 1975, Avila and Bergström 2006/
<i>IR_{Water}</i> Intake rate of water	m³/y	4.8	0.6		/ICRP 1975, Avila and Bergström 2006/

(*) For the lognormal (LGN) and normal (N) distributions the first parameter is the Mean and the second the Standard Deviation of the untransformed data. For the triangular distribution (Triang) the first parameter is the minimum, the second the Maximum and the third the Mode. For the uniform distribution (Unif) the first parameter is the Minimum and the second the Maximum.

The value of the Dose Conversion Coefficient for food ingestion ($DCC_{ingFood}$) used for calculations of food ingestion doses (Equation 4.6) was taken from /Avila and Bergström 2006/, and is the value recommended in /ICRP 1996/. The value of the Dose Conversion Coefficient ($DCC_{ingWater}$) used for calculations of doses from water ingestion (Equation 4.8) was taken from /Leggett 2004/.

 $DCC_{ingFood} = 5.8.10^{-10} \text{ Sv/Bq}$ $DCC_{ingWater} = 2.9.10^{-11} \text{ Sv/Bq}$

The appropriate averaging period (T_{av}) will depend on the period over which the annual doses are averaged. For instance, if the average lifetime annual dose for an adult person is to be estimated then an averaging period of 50 y is appropriate, which is the integration period used in /ICRP 1996/ for derivation of the DCC for adults.

4.4 Sensitivity and uncertainty analyses

A sensitivity analysis was carried out to study the effect on the model predictions of variations in the model parameters. An uncertainty analysis was also carried out to assess how parameter uncertainties propagate through the models, resulting in uncertainties in the model predictions. Both analyses were carried out with the software package Eikos /Ekström and Broed 2006/ by performing probabilistic simulations (10,000 samples) using Latin Hypercube sampling. As sensitivity index the Spearman Rank Correlation Coefficients (SRCC) was used, which is an appropriate sensitivity measure for the models studied herein, with monotonic relationships between parameters and outputs. Three cases were considered in the sensitivity study, where for parameters that are usually easy to obtain for each specific situation, such as the area of the sea basin, the lake and the forest ecosystem growing on lake sediments, the values were taken from the SFR 1 SAR-08 /SKB 2008/ safety assessment.

The three cases considered were:

- 1. Continuous unit release rate (1 Bq/y) to a sea basin with an area of 1.4E7 m² and an average depth of 9.5 m with values ranging from 8.5 to 10 m (a triangular distribution was assigned).
- 2. Continuous unit release rate (1 Bq/y) to a lake with an area of 1.6E6 m² and a catchment area of 1.4E7 m².
- 3. Exposure in a forest ecosystem growing on the sediments of the lake used in Case 2. For the forest an area of 1.4E6 m² was assumed, which is smaller than the area of the lake, but nevertheless it was conservatively assumed that the whole ¹⁴C inventory in the lake sediment was transferred to the forest soil. A triangular distribution was assigned to the parameter T_{release} (duration of the release to the lake in years) with parameters: Min: 1,000, Max: 3,000 and Mode: 2,000. These are typical times required for transformation of lakes into mires in the areas of interest /SKB 2006/. The annual doses from the contaminated forest were estimated using the model described in Chapter 3, with release rates calculated with Equation 4.5.

For all other parameters the PDFs, or nominal values, where PDFs were missing, given in Tables 3-1 and 4-1, were used in the simulations.

Statistics of the total annual doses per unit release rate (annual doses from ingestion of food and inhalation) obtained for the three studied cases are presented in Table 4-2. The dose rate values for the lake are three orders of magnitude higher than the ones for the sea basin. This can be explained by the smaller area of the lake and the substantially lower residence time of the released ¹⁴C in the sea basin. For both cases, the ratio between the 95 and 5 percentiles is below 10. For the case of a forest growing on contaminated lake sediments, the annual doses from the forest are substantially lower that the annual doses from the lake. This indicates that exposure to ¹⁴C accumulated in lake or sea sediments is a less important exposure pathway for humans, as compared to the exposure from the lake or sea basin in the release period.

Table 4-2. Statistics of the total annual doses (in Sv/y) obtained for the three calculation cases with continuous releases of 1 Bq/y to a sea basin, a lake and for the case of a forest growing on contaminated lake sediment.

Case	Mean	SD	5%	50%	95%
Sea basin	1.7E–16	5.8E-16	5.0E-17	1.1E–16	3.7E–16
Lake	1.7E–13	2.2E-14	1.4E–13	1.7E–13	2.1E–13
Forest on lake sediment	3.0E-15	5.9E–15	4.6E-16	1.7E–15	8.5E–15

The contribution of ingestion of food and water to the total annual doses per unit release rate to the lake are shown in Table 4-3. It can be appreciated that the food ingestion doses are several orders of magnitude higher than the doses from ingestion of water.

Statistics of the excess ${}^{14}C/{}^{12}C$ ratios (specific activity) obtained for the three studied cases are presented in Table 4-4. The relationship between the specific activities obtained for the three cases is the same as the relationship between the annual doses (see Table 4-2) and the observed differences have the same explanation as given above for the doses.

Results of the sensitivity study are shown in Figures 4-2 and 4-3. There is a marked difference between the three studied cases regarding how predicted annual doses per unit release rate are influenced by variations in the model parameters. For the case of releases to the sea basin, the dissolved inorganic carbon (DIC) and the residence time (RT) have a dominant effect on the dose predictions. This is due to the high rate of exchange of the water in comparison with the rate of carbon assimilation by primary producers. However, for the case of releases to the lake there is a similar effect of parameters related to the water exchange (DIC) and those related to the assimilation rate of carbon by primary producers (NPP). In the case of the forest growing on contaminated lake sediments the wind speed is the parameter with the largest effect on the predicted doses. The next most influencing parameter in this case is the decomposition rate of the organic matter, which determines the release rate of ¹⁴C to the mixing layer, where it can be assimilated by the canopy.

•	Table 4-3.	Mean values	of the annual	doses from	food ingesti	on and water	ingestion	
((in Sv/y) c	obtained for t	he calculation of	cases with c	ontinuous r	eleases of 1 E	3q/y to a lake	

Case	Food ingestion	Water ingestion
Lake	1.7E–13	1.0E-18

Table 4-4. Statistics of the excess ${}^{14}C/{}^{12}C$ Ratios (specific activity in Bq/g C) obtained for the three studied cases with continuous releases of 1 Bq/y to a sea basin, a lake and for the case of a forest growing on contaminated lake sediment.

Case	Mean	SD	5%	50%	95%
Sea basin	2.7E-12	9.1E–12	7.8E–13	1.7E–12	5.8E-12
Lake	2.7E-9	3.4E-10	2.2E-9	2.7E-9	3.3E–9
Forest on lake sediment	4.4E-11	8.9E-11	6.9E-12	2.5E-11	1.3E–10



Figure 4-2. Spearman Rank Correlation Coefficients (SRCC) of the total ${}^{14}C$ annual doses per unit release rate obtained for the cases of release to a sea basin (left figure) and a lake (right figure). The parameter names can be found in Table 4-1.



Figure 4-3. Spearman Rank Correlation Coefficients (SRCC) of the total ¹⁴C annual doses per unit release rate obtained for the case of exposure on a forest growing on contaminated lake sediments. The parameter names can be found in Tables 3-1 and 4-1.

5 Conclusions

The models proposed in this report can be used in assessments of the exposures resulting from releases from the geosphere to various ecosystem types, including terrestrial ecosystems, like forests and agricultural lands, as well as aquatic ecosystems, such as sea basins and lakes. A model for assessment of exposures resulting from irrigation with contaminated water, for example with water from a contaminated well is also included. The proposed models are based on the so-called specific activity approach /Killough 1980/, which has been recommended by the /UNSCEAR 2000/ and /IAEA 2001/ for assessment of doses resulting from ¹⁴C releases to the environment from nuclear installations, such as nuclear power plants. In deriving the equations for estimation of ¹⁴C specific activities in environmental compartments several realistic and conservative assumptions were introduced. These assumptions are documented and justified in Chapters 2, 3 and 4. The models can be used in safety assessments of geological repositories of radioactive waste, to carry out cautious but still not over conservative dose estimations, which can be compared with regulatory dose constrains.

The comparative analyses carried out with the models in Chapters 3 and 4 indicate that the worse situations will be associated with releases to small lakes, especially if these have a low productivity and relatively long water residence times, as compared with the characteristic time of the processes of carbon assimilation by primary producers. Depending on the dose levels obtained in the assessments, it might be necessary to reduce the conservatism of the models. This could be achieved by developing dynamic models, as the ones proposed in /SKB 2004b/ for coastal ecosystems and running these until equilibrium is achieved.

Releases to terrestrial ecosystems and sea basins will generally result in lower doses, as in these systems the residence time of the released ¹⁴C in air and water, respectively, is very short compared with the characteristic time of the carbon assimilation by primary producers. The study also indicates that accumulation of ¹⁴C in sediments and subsequent exposure to terrestrial systems formed on these sediments does not result in substantial doses, as compared with doses received from releases to the corresponding aquatic ecosystem.

In all cases considered in this study, the doses via food ingestion were much higher than the doses by inhalation and water ingestion, which is consistent with results reported in other studies /UNSCEAR 2000/.

The uncertainty analysis shows that for all cases, except for the case of irrigation, the dose predictions are within a factor of 5 to 20 (from the 5 to the 95 percentiles of the values). For the case of irrigation a larger variation (within a factor of 200) of the predicted doses was observed, but this was due to a large variation assumed for the well capacity. It should be noted that a larger uncertainty in the dose estimations is obtained if calculations are done using transfer factors, as these show a variability of several orders of magnitude. When applying the models proposed herein, it should be considered that several conservative assumptions have been made to deal with some uncertainties. For example, the uncertainty in the fraction of the released ¹⁴C that is available for intake by photosynthesis has been "eradicated" by assuming that 100% of the release is in a form that can be readily taken up by plants.

Results of the sensitivity analysis indicate that few parameters have a dominant effect on the dose predictions. These are parameters that are relatively easy to measure, such as the wind speed, the concentration in water of dissolved inorganic carbon (DIC), the water residence time (RT), the net primary production (NPP) (see Sections 3.4 and 4.4). One parameter that may be difficult to assign values to is the area of the contaminated object (release area), as the values will depend on transport processes in the bedrock and regolith. However, values given to the area will not only affect the doses – the smaller the area the larger the doses, but also the number

of exposed individuals – the smaller the area the smaller the number of exposed individuals. For example, if the area of the agricultural land in the case considered in Section 3.4 is reduced to $1,000 \text{ m}^2$ the doses will increase to around 5.5E-15 Sv/y, but only one individual could receive such dose (using the values of productivity of agricultural lands reported in /Lindborg (ed) 2005/ for the Forsmark area), as compared to as many as 1,600 individuals that would receive the dose that is obtained if a larger area is assumed, as in Section 3.4.

6 References

Avila R, Bergström, U, 2006. Methodology for calculation of doses to man and implementation in Pandora. SKB R-06-68, Svensk Kärnbränslehantering AB.

Avila R, Thiry Y, Gilbin R, Agüero A, Thorne M, Sheppard M, Tamponnet C, Ikonen A, Xu S, 2006. Recommendations for improving predictions of the long-term environmental behaviour of ¹⁴C, ³⁶Cl, ⁹⁹Tc, ²³⁷Np and ²³⁸U; Findings of the IUR "Radioecology and Waste" Task Force IUR Report 6, 2006 (http://www.iur-uir.org/upload/xeyyqtjqie.pdf).

Bergström U, Barkefors C, 2004. Irrigation in dose assessments models. SKB R-04-26, Svensk Kärnbränslehantering AB.

Boeckx P, van Cleemput O, 1996. Influence of chemical and physical soil properties on CH₄ oxidation in soils. In: P. Borrell, P. Borell, T. Cvitas, K. Kelly, W. Seiler (ed).: Transport and Transformation of Pollutants in the Troposphere; Vol. 2, 409–413, Computational Mechanics Publications, Southampton, 1996.

Casper P, Maberly S C, Hall G H, Finlay B J, 2000. Fluxes of methane and carbon dioxide from a small productive lake to the atmosphere. Biogeochemistry, 19: 1–19.

Casper P, Chan C O, Furtado A L S, Adams D D, 2003. Methane in an acidic bog lake: The influence of peat in the catchment on biogeochemistry of methane. Aquat. Science, 65: 36–46.

Ekström P-A, Broed R, 2006. Sensitivity Analysis Methods and a Biosphere. Test Case Implemented in Eikos. Posiva Working Report 2006-31.

Engqvist A, Andrejev O, 1999. Water exchange of Öregrundsgrepen. A baroclinic 3D-model study. SKB TR-99-11, Svensk Kärnbränslehantering AB.

Geisler G, 1980. Pflanzenbau; Verlag Paul Parey. Berlin und Hamburg, 1980.

Gisi U, 1990. Bodenökologie Georg Thieme Verlag, Stuttgart and New York, 1990.

Grünhage L, Jäger H, 2001. Austausch von Stoffen zwischen Atmosphäre und Biosphäre. In: Guderian, R. (ed).: Handbuch von Umweltveränderungen und Ökotoxikologie, Band 2a. Terrestrische Ökosysteme: Immissionsökologische Grundlagen – Wirkungen auf Boden – Wirkungen auf Pflanzen, Springer Verlag Berlin, 2001.

Grünhage I, Hanewald K, Jäger H, Ott W, 1999. Auswirkung dynamischer Veränderungen der Luftzusammensetzung und des Klimas auf terrestrische Ökosysteme in Hessen.

Heilmann M A, Carlton R G, 2001. Methane oxidation associated with submersed vascular macrophytes and its impact on plant diffusive processes. Biogeochemistry, 52: 207–224.

IAEA, 2001. Generic models for use in assessing the impact of discharges of radioactive substances to the environment. Safety Reports Series No. 19, 2001.

ICRP, 1975. Reference man: anatomical, physiological, and metabolic characteristics. Publication 23, Pergamon Press, Oxford.

ICRP, **1996.** Age-dependent doses to members of the public from intake of radionuclides: ICRP Publication 71 and 72. 1996.

Killough G G, 1980. A dynamic model for estimating radiation dose to the world population from releases of 14C to the atmosphere. Health Physics 38: 269–300.

Kumblad L, Gilek M, Naeslund B, Kautsky U, 2003. An ecosystem model of the environmental transport and fate of carbon-14 in a bay of the Baltic sea, Sweden. Ecological modeling 166: 193–210.

Leggett R W, 2004. A biokinetic model for carbon dioxide and bicarbonate. Radiation Protection Dosimetry, 108: 203–213.

Lindborg T (ed), 2005. Description of surface systems. Preliminary site description Forsmark area – version 1.2. SKB R-05-03, Svensk Kärnbränslehantering AB.

Lindgren M, Pettersson M, Karlsson S, Moreno L, 2001. Project SAFE Radionuclide release and dose from the SFR repository. SKB R-01-18, Svensk Kärnbränslehantering AB.

Mayall A, 2003. Modelling the dispersion of radionuclides in the atmosphere; In: M. Scott (ed); Modelling of Radioactivity in the environment, Elsevier, 2003.

Rudd W M, Hamilton R D, 1978. Methane cycling in an eutrophic shield lake and its effects on whole lake metabolism. Limnology and Oceanography, 23: 337.

Schmaljohann R, 1996. Methane dynamics in the sediment and water column of Kiel Harbour (Baltic Sea). Mar. Ecol. Prog. Ser. (131): 263–273.

Sheppard S C, Ciffroy P, Siclet F, Damois C, Sheppard M I, Stephenson M, 2006a. Conceptual approaches for the development of dynamic specific activity models of ¹⁴C transfer from surface water to humans, J. Environ. Radioactivity, 87: 32–51.

Sheppard S C, Sheppard M I, Siclet F, 2006b. Parameterization of a dynamic specific activity model of ¹⁴C transfer from surface water-to-humans, J. Environ. Radioactivity, 87: 15–31.

Seinfeld J, 1986. Atmospheric chemistry and physics of air pollution; Wiley interscience Publication, New York.

SKB, **2003.** Shore displacement in northern Uppland during the last 6500 calender years. SKB TR-03-17, Svensk Kärnbränslehantering AB.

SKB, **2004a**. Investigation of marine and lacustrine sediment in lakes. Stratigraphical and analytical data. Forsmark site investigation. SKB P-04-86, Sv. Kärnbränslehantering AB.

SKB, 2004b. Models for transport and fate of carbon, nutrients and point source released radionuclides to an aquatic ecosystem. SKB TR-04-13, Svensk Kärnbränslehantering AB.

SKB, 2006. The Biosphere at Forsmark. SR-Can. SKB R-06-82, Svensk Kärnbränslehantering AB.

SKB, 2007. Collection of archive samples. Forsmark site investigation. SKB P-07-196, Svensk Kärnbränslehantering AB.

SKB, 2008. SFR 1 SAR – Allmän del 2 – Analys av förvarets långsiktiga säkerhet, Svensk Kärnbränslehantering AB.

Smith P, Neall F, Snellman M, Pastina B, Nordman H, Johnson L, Hjerpe T, 2007. Safety assessment for a KBS-3H spent nuclear fuel repository at Olkiluoto – Summary report, POSIVA 2007-06. Posiva Oy, Olkiluoto, Finland.

Strasburger E, 2002. Lehrbuch der Botanik (Textbook of Botany), Spektrum Akademischer Verlag, 2002.

Troen R, Lundtang Petersen E, 1991. European Wind Atlas, Risoe National Laboratory, Risoe, Denmark, 1991, ISBN 87-550-1482-8.

UNSCEAR, 2000. Sources and Effects of ionizing radiation. United Nations Scientific Committee on the Effects of Atomic Radiation. Volume 1. United Nations.

Utsumi M, Nojiri Y, Nakamura T, Nozawa T, Otsuki A, Seki H, 1998. Oxidation of dissolved methane in eutrophic, shallow lake: Lake Kasumigaura, Japan. Limnology and Oceanography, 43: 471–480.

Venkiteswaran J J, Schiff S L, 2005. Methane oxidation, isotopic enrichment factors in freshwater boreal reservoirs. Applied Geochemistry; 20: 683–690.