

# **Prototype dynamic tool for the assessment of radiological exposure to non-human biota in a coastal heath dune ecosystem**

By

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## Abstract

The objective of this work is a proof-of-concept dynamic assessment model for a terrestrial environment based on simple, relatively well known parameters. The model has been developed to reproduce trends of  $^{137}\text{Cs}$  in a coastal heath dune ecosystem. This study has provided a useful tool for further research in radioecology as it can be further developed to include more radionuclides and calibrated to different ecosystems. Model output has shown that the approach developed here for simulating radionuclide turnover by biota can reproduce realistic trends of  $^{137}\text{Cs}$  in a semi-natural environment. The model includes the capability of calculating the dose to vegetation from externally deposited radionuclides. This is a novel calculation that allows a more complete assessment of the risk to flora from radionuclides that are released by anthropogenic activities to the environment. The key aspects of the dynamic modelling approach are the calculation of an uptake rate by biota as a function of relatively well known parameters available in the scientific literature: the Concentration Ratio, biological half-life, and mass values for the organism and soil.

The modelling approach uses the software package Modelmaker 4.0 to construct a compartmental model with flows between compartments to simulate the flow of  $^{137}\text{Cs}$  between different physical and biological components of the environment and to calculate the absorbed dose received by biota. The flow of  $^{137}\text{Cs}$  between physical compartments is modelled using empirical formula derived from the scientific literature with deposition from the atmosphere modelled using a simple lumped parameter of the deposition velocity of  $^{137}\text{Cs}$ . The migration of  $^{137}\text{Cs}$  from the top 0.1 m of soil is simulated using a simple clearance model that derives the migration rate of  $^{137}\text{Cs}$  based on empirical measurements from a sand dune ecosystem. Biota are modelled using a reference animal and plant approach with data from the scientific literature used to model a generic detritivore, omnivore, carnivore, herbivore, and plant that have parameters and life histories resembling those in biota commonly found in heath dune ecosystems in the UK. An initial warm-up run was carried out to represent the conditions at Drigg dunes in the years 1990-1995 using empirical data for atmospheric concentration of  $^{137}\text{Cs}$  at Drigg at this time. This allowed sufficient time for the clean biota compartments to reach the level of  $^{137}\text{Cs}$  concentration prevailing in biota prior to a hypothetical accident release scenario. A second scenario is run to simulate the model results from a hypothetical accidental release of  $^{137}\text{Cs}$ . This is modelled using the deposition data of  $^{137}\text{Cs}$  recorded at the Drigg dunes from the Chernobyl incident. This data is used to model a hypothetical accident from the years 1996-2000.

The dose hierarchy is as follows for the warm-up scenario: Detritivore > Vegetation > Omnivore > Carnivore > Herbivore. For the warm-up scenario, the model showed that the highest dose received by vegetation is  $0.08 \mu\text{Gy hr}^{-1}$  within a few days after initial contamination and a dose of  $0.06 \mu\text{Gy hr}^{-1}$  by the end of the simulation period. The highest dose received by the detritivore was  $0.09 \mu\text{Gy hr}^{-1}$ . The estimated absorbed dose by biota did not follow the same trend as the organism activity concentrations. The soil  $^{137}\text{Cs}$  concentration has a larger effect on the absorbed dose and so occupancy factors were important in determining the absorbed dose.

Sensitivity analysis shows model output for activity concentration is most sensitive to the Concentration Ratio parameter as varying this parameter has a linear effect on model output for herbivore activity concentration. The next most sensitive parameter tested was the biohalf-life where a doubling of this parameter reduced herbivore activity concentration by just under 35% while the least significant parameter analysed was the soil migration rate where a doubling of this parameter decreased herbivore activity concentration by 20% compared to the default migration rate.

The reference vegetation was modelled with a separate external and internal compartment to allow the effect of deposited  $^{137}\text{Cs}$  from the atmosphere and  $^{137}\text{Cs}$  taken up by vegetation roots from the soil to be accurately simulated as separate processes. The significance of including an external vegetation compartment varied between the emergency and existing exposure scenarios. An external vegetation compartment had very little impact on the vegetation activity concentration for the warm-up scenario which had a low deposition rate. However, under the emergency scenario the externally deposited  $^{137}\text{Cs}$  caused a much larger spike in vegetation activity concentration in comparison with the spike seen in the reference animal biota. The vegetation compartment reaches a peak of  $3\text{E}04 \text{ Bq kg}^{-1}$  for the emergency scenario, while the highest activity concentration reached by the animal biota is the herbivore at  $14 \text{ Bq kg}^{-1}$ . An independent data set from a heath dune ecosystem is required for model validation.

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## Variable notation list

Variable	Symbol	Units
Above ground biomass	$B$	kg
Activity	$A$	Bq
Activity per unit surface area	$\zeta$	Bq m <sup>-2</sup>
Activity concentration	$C$	Bq per unit mass (kg) or volume (m <sup>3</sup> )
Biological halflife	$T_{B1/2}$	d
Decay constant	$\lambda$	d <sup>-1</sup>
Density	$\rho$	kg m <sup>-3</sup>
Deposition rate	$D$	Bq d <sup>-1</sup>
Deposition rate per unit surface area	$\zeta$	Bq m <sup>-2</sup> d <sup>-1</sup>
Deposition velocity	$v_d$	m d <sup>-1</sup>
Distribution coefficient	$K_d$	L kg <sup>-1</sup>
Dose to biota	$Ds$	Gy
Dose per unit concentration	$DPUC$	(Gy h <sup>-1</sup> )/(Bq kg <sup>-1</sup> )
Effective release rate	$k$	d <sup>-1</sup>
Environmental halftime	$T_w$	d
Expected lifetime of organism	$E_{life}$	d
Interception coefficient	$\mu$	m <sup>2</sup> kg <sup>-1</sup>
Interception factor	$\varphi$	unitless
Mass	$M$	kg
Migration rate	$r$	d <sup>-1</sup>
Number of atoms	$N$	N/A
Number of stomata per leaf surface area	$n$	m <sup>-2</sup>
Physical halflife	$T_{1/2}$	years or days
Porosity	$\omega$	unitless
Rainfall rate	$R$	m y <sup>-1</sup>
Release rate	$RI$	d <sup>-1</sup>
Scavenging coefficient	$\Lambda$	s <sup>-1</sup>
Soil depth	$\delta$	m
Surface area	$S$	m <sup>2</sup>
Translocation rate	$TR$	m <sup>2</sup> d <sup>-1</sup>
Uptake rate	$U$	d <sup>-1</sup>
Washout rate	$w$	d <sup>-1</sup>
Weathering rate	$WR$	d <sup>-1</sup>

Width of epidermis	$l_s$	m
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# Introduction

Until the early 1990s, the main guidance from the International Commission on Radiological Protection (ICRP) recommended the standard of environmental control needed to protect humans will ensure that other species are not put at risk (ICRP, 1991). Thompson (1988) showed there was a lack of supporting scientific data to back the ICRP's assumption that protecting humans will also protect top marine predators in the UK. Pentreath (1998) showed no internationally agreed criteria for protecting the environment from the effects of ionising radiation existed. The ICRP has since changed its stance on environmental protection and a number of different assessment tools have been developed to assess the radiological exposure to non-human biota following radionuclide releases from anthropogenic activities.

An established international framework for the protection of the environment from radiation exposure is still currently an evolving subject (Pentreath, 2009). The effort to establish a system parallel to the protection of humans has been motivated by the need to fill a conceptual gap in radioprotection rather than a response to any specific environmental issue. The ICRP have published recommendations that include an approach for developing a framework to demonstrate radiological protection of the environment (ICRP, 2007). The recommendations also move from the previous approach on radiological safety based on practices and interventions to an approach based on three exposure situations of existing, planned, and emergency situations (ICRP, 2007). These recommendations have been developed by the ICRP, most notably with the publication of their Reference Animal and Plants (RAP) report, which introduces the concept of Reference Animals and Plants, and defines a small set (ICRP, 2009).

The main pathways for radionuclides in the environment are outlined in Wood et al. (2009b). For the physical environment these are deposition from the atmosphere on to vegetation or soil and the subsequent removal of radionuclides from the vegetation surface or sequestering of radionuclides through the plant stomata. Radionuclides deposited onto the soil will either be resuspended into the atmosphere or migrate through various processes such as leaching by water or dispersion through Brownian motion into deeper areas of the soil or into groundwater. Radionuclides may also be incorporated or fixed into the soil by binding to certain parts of the soil matrix or enter the biological food-chain through various pathways such as ingestion or uptake by plant roots. The methodologies used to estimate radionuclide concentrations in biota are key in the protection of biota from ionising radiation.

There are a number of methodologies for assessing the activity concentration in non-human biota. This can be estimated by using empirically derived Concentration Ratios (CRs).  $CR = \text{activity concentration in organism (Bq kg}^{-1}) / \text{activity concentration in environment (Bq kg}^{-1})$ . If the activity in an environmental medium is known, the activity in an organism can be estimated and the dose to biota calculated, provided a sufficiently accurate Concentration Ratio is available from the scientific literature. Various assessment tools have been developed using the Concentration Ratio approach to allow the risk to the environment from radiological activities to be calculated such as Copplestone et al. (2001) and ERICA (Wood et al., 2009a) although their use in industry radioprotection is debated (Brownless, 2007).

When there is a pulse of radionuclides released to the environment, the radioactivity concentration of soil and biota may no longer be in equilibrium. If the residence time of the radionuclides in the environment is shorter than the biological half-life there is no longer equilibrium of activity between the environment and organisms. This means the use of Concentration Ratios will not be as accurate in estimating the activity concentration in biota and will result in an over-prediction in the biota dose assessment. This over-prediction is a method favoured by regulators as it means they are more confident the assessed environmental risk is conservative and erring on the side of caution.

Modelling assessments carried out in a marine environment by Vives i Batlle et al. (2008) showed that dose assessment from dynamic models on a time scale below 2 years can be considerably different to using a model that assumes equilibrium (Fig. 1). In Figure 1 there is a difference in the dynamic model output for the winkle compared to the macrophyte. This is due to the macrophyte being modelled with a biphasic release of internal radionuclides with a short initial release and a second slower release over longer time-periods while the winkle only has a single long-term phase (Vives et al. 2008).

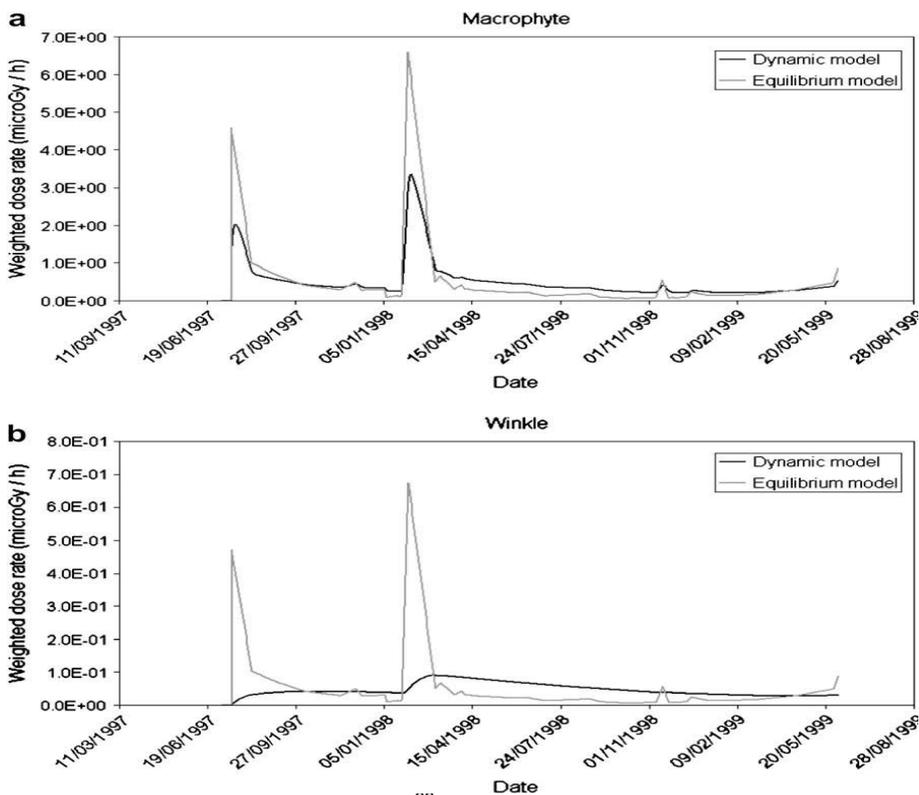


Figure 1: Predicted dose rates for  $^{99}\text{Tc}$  in macrophyte and winkles in the Drigg area (taken with permission from Vives i Batlle et al., 2008).

Under its statute, the International Atomic Energy Agency (IAEA) has the specific mandate to establish, in consultation and collaboration with member states and specialized agencies, standards for protection against ionising radiation. The IAEA's comprehensive standard of protection for all activities that involve radioactive substances is the International Basic Safety Standards (BSS). The BSS, last published in 1996 and currently under revision, expand and interpret the ICRP recommendations in practical terms while also

considering the findings of other scientific organizations such as the United Nations Scientific Committee on the Effect of Atomic Radiation (UNSCEAR). The EMRAS II project run by the IAEA aims to improve environmental modelling methods for assessing dose to biota through improving and exchanging data and models. A number of technical documents based on the findings of working groups will be produced. The Biota Modelling Group part of EMRAS aims to compare and validate models being used or developed for biota dose assessment as part of regulatory compliance monitoring of authorized planned releases. Providing an approach that is based on a few, well-known, simple parameters allows the feasibility of dynamic modelling for terrestrial ecosystems and its accuracy to be investigated.

Legal requirements for investigating the environmental impact of radionuclides released from anthropogenic activities include the European Habitats and Wild Birds Directive which requires the impact of ionising radiation on Natura 2000 sites to be assessed. The Water Framework Directive includes the need to assess the impact of a range of chemicals including radionuclides as a carcinogen. The ICRP's framework for describing situations in which exposure to radiation may occur has three scenarios: planned, existing and, emergency. The emergency situation is particularly likely to produce non-equilibrium conditions for which a dynamic modelling approach is more appropriate for assessing radiological risk.

Dune ecosystems provide a good case study within the UK for assessing radiological risk as many of these sites are protected habitats or contain protected species. The Drigg coastal dunes near the Sellafield nuclear site are part of the Drigg Coast Natura 2000 site and contain the protected species natterjack toad (*Bufo calamita*) and great crested newt (*Triturus cristatus*). Also, many coastal ecosystems are in close proximity to the UK's nuclear facilities. The dune environments at Drigg (Wood et al., 2008) and at other dune environments near Sellafield (Wood et al., 2009b) have had empirical data collected from them so there is a database of information available for parameters in a prototype dynamic assessment model.

## **Objectives**

The primary objectives of this study are to:

1. Produce a proof-of-concept dynamic model for assessing the environmental risk to terrestrial environments from ionising radiation.
2. Analyse the results of the prototype assessment tool for calculating biota activity concentration and absorbed dose to biota from  $^{137}\text{Cs}$  in a coastal heath dune ecosystem for an exposure situation involving low levels of  $^{137}\text{Cs}$  atmospheric concentration.
3. Analyse the results of the prototype assessment tool for calculating biota activity concentration in a coastal heath dune ecosystem for an emergency exposure situation involving relatively high levels of  $^{137}\text{Cs}$  atmospheric concentration.
4. Perform a sensitivity analysis on key model parameters: the Concentration Ratio, the biological half-life, and the soil migration rate for their affect on herbivore activity concentration.
5. Perform a sensitivity analysis on the weathering rate for the affect on vegetation activity concentration.

# Methods

## *Model design*

### *Model overview*

Simulating the risk to biota in this study is carried out using the Modelmaker 4.0 software. This allows the user to create compartments that represent different components of the sand dune ecosystem. Arrows from the different compartments allow the changing concentration of radioactivity to be dynamically modelled over a given time period. So, for example, the radioactivity, or activity,  $A$ , in a compartment (in units of Bq) can be calculated as  $A = N\lambda$  where  $N$  = number of atoms,  $\lambda = \frac{\ln(2)}{T_{1/2}}$  is the decay constant ( $d^{-1}$ ),  $\ln(2) = 0.693$  and  $T_{1/2}$  is the physical half-life of the radionuclide. From every compartment there is an outgoing flow representing the decay of the radionuclide. The decay rate can be entered in Modelmaker as  $\frac{dA}{dt} = -\lambda A$ .

Modelmaker is designed primarily for simulating a number of sub-processes that can be combined to represent the behaviour of a larger more complicated system and so is appropriate for the design of a biokinetic model. The user interface of Modelmaker provides a schematic of the completed dynamic model (Fig. 3) and how various sub-processes are combined to model the dynamics of activity concentration for a terrestrial environment.

It is the radionuclides in a compartment which are the source of the activity. A number of different pathways can contribute to the uptake of radionuclides by terrestrial biota. These pathways cover a range of different processes such as radionuclide transport in the atmosphere, environmental contamination through deposition to the soil and the actual uptake of radionuclides by biota. The importance of different pathways can change over time after the initial release of radionuclides from an accident event. In an emergency scenario, the initial plume or shine path and initial uptake processes such as inhalation will be significant pathways to consider for absorbed dose. The initial environmental contamination will come from the deposition of radionuclides from the atmosphere to vegetation or the soil. As the period of time from the initial release of radionuclides increases the important pathways to consider will be those that affect the change of radionuclide concentration in different soil compartments such as the rooting zone and how this affects the concentration of radionuclides within biota. The key uptake parameter in the modelling approach developed here is the Concentration Ratio. This relates the concentration of activity in biota to the concentration of activity in the top 0.1 m of soil. Therefore, it is the pathways that will have a significant effect on radionuclide concentration in the top 0.1 m of soil that is considered in the dynamic model developed in this study. Wood et al. (2009b) provides a conceptual overview of the main nuclide fluxes in a sand dune ecosystem (Fig. 2), which provides the conceptual basis of the prototype dynamic model developed here.

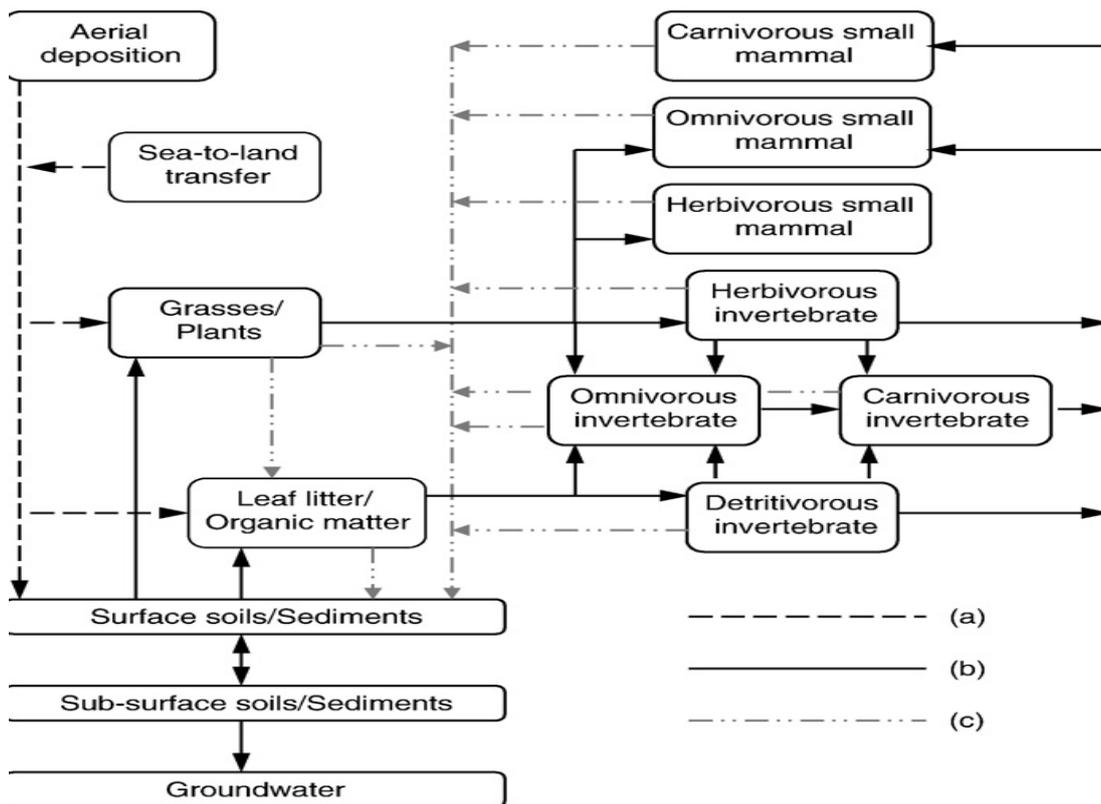


Figure 2: Conceptual model of the main radionuclide fluxes in a coastal sand dune ecosystem near the Drigg dunes ecosystem. (a) is the flux of anthropogenic radionuclides (b) uptake by biota (c) routes of return from biota to the environment through excretion, mortality etc. Arrows indicate the direction of flow for radionuclides. Diagram from Wood et al. (2009b).

In this study the model is only used for the radionuclide  $^{137}\text{Cs}$ . This is due to the model being at the proof-of-concept stage so the demonstration of model output for only one radionuclide that is likely to have a relatively large amount of data availability in the scientific literature is sufficient. The model is run using annual parameters and data averaged over 12 monthly time steps. This is appropriate for medium to long lived radionuclides that have a half-life consisting of many years. For shorter lived radionuclides the model would have to be adapted to include accurate sub-annual data for processes such as variation in deposition rate of radionuclides from the atmosphere caused by changes in rainfall levels.



Figure 3: Modelmaker user interface and schematic of the biokinetic model for the uptake and release of  $^{137}\text{Cs}$  by non-human biota developed for this study. Physical processes of atmospheric deposition, soil migration, and weathering of  $^{137}\text{Cs}$  from external vegetation are fixed parameters as is the translocation of  $^{137}\text{Cs}$  from the external vegetation to the internal vegetation via stomatal uptake. The dynamic pathways assessed by this study are internal uptake and release by biota. It is the dynamic biological pathways that this study is primarily focussed on reproducing.

## Biological turnover of radionuclides

In radioecology the uptake of radionuclides by animals and plants in an environment can be represented by two well-known parameters:

$$CR = \frac{C_o}{C_s} \quad (1)$$

where:

$CR$  = Concentration Ratio

$C_o$  = Activity concentration of organism ( $\text{Bq kg}^{-1}$ )

$C_s$  = Activity concentration of soil ( $\text{Bq kg}^{-1}$ )

The Concentration Ratio is an equilibrium parameter that can be used for this study where the focus is on internal pathways. There is a simple way to put the Concentration Ratio as a function of the number of atoms in organism and soil ( $N_o$  and  $N_s$ ) when the system has reached equilibrium, as well as the total mass of soil and organism,  $M_s$  and  $M_o$ , using the equation  $A = N\lambda$  where:

$$CR = \frac{N_o \times \lambda / M_o}{N_s \times \lambda / M_s} = \frac{N_o}{N_s} \times \frac{M_s}{M_o} \quad (2)$$

The second key quantity is the biological half-life ( $T_{B1/2}$ ), which is the time it takes for 50% of radionuclides taken up by an organism to be released from the organism through biological processes. This process is symbolised as:

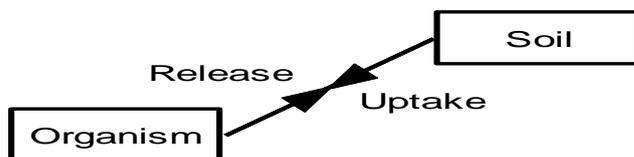


Figure 4: Schematic representation in Modelmaker of uptake and release of radionuclides by biota.

There is a simple way to calculate the uptake and release rates as a function of the Concentration Ratio and  $T_{B1/2}$ , by assuming equilibrium which is when the uptake and release flows are equal, so that:

$$U \times N_s = Rl \times N_o \quad (3)$$

where:

$U$  = uptake rate ( $d^{-1}$ )

$Rl$  = release rate ( $d^{-1}$ )

$$Rl = \frac{\ln(2)}{T_{B1/2}} \quad (4)$$

$$U = Rl \times \frac{N_o}{N_s} = \frac{\ln(2)}{T_{B1/2}} \frac{N_o}{N_s} \quad (5)$$

The ratio  $\frac{N_o}{N_s}$  at equilibrium can be put as a function of the Concentration Ratio and the masses using

$$\frac{N_o}{N_s} = CR \frac{M_o}{M_s} \quad (\text{from Eq. [2]}):$$

$$U = \frac{\ln(2)}{T_{B1/2}} CR \frac{M_o}{M_s} \quad (6)$$

So, with equations [4] and [6] simple dynamic uptake and turnover of radionuclides by biota can be modelled as a function of two parameters (Concentration Ratio and  $T_{B1/2}$ ), which are commonly found in the literature, plus the mass values for both the organism and the soil compartment.

### *Radionuclide loss from vegetation*

In addition to the processes described above the vegetation compartment in the model has additional uptake and loss processes. Loss of externally deposited radionuclides is estimated using a weathering rate. This is calculated using a weathering constant of 4.22 (derived from empirical evidence in IAEA, 1996) and multiplying by the activity concentration in the vegetation external compartment. The weathering rate is calculated using the environmental half-time of a radionuclide. This is the time necessary for one-half of the activity to be removed from a certain part of the environment of interest, such as the vegetation, by environmental processes. The formula used is:

$$WR = \frac{\ln(2)}{T_w} \quad (7)$$

where:

$WR$  = the weathering rate ( $d^{-1}$ )

$T_w$  = environmental half-time (d)

An environmental half-time is taken from the review of data in IAEA (1996) which reports a range of environmental half-times reported in the literature for  $^{137}\text{Cs}$ . A range of values from 40-60 days are reported over what the IAEA considers as long-term (IAEA, 1996) which is a period of at least 2 months. As long-term data wasn't available for grassland, the environmental half-time for pasture was used. As the longest reported half-time of 60 days is used here, the derived weathering rate is considered conservative for leafy vegetation.

The additional uptake process is translocation. The radionuclides are transferred from the external part of the plant leaf to the internal part of the plant leave via the stomata, which is referred to as translocation. The translocation rate,  $TR$  ( $m^2 d^{-1}$ ), of washed-out particles back into the plant is assumed to be

$$TR = wS_oS_l \quad (8)$$

where:

$S_o$  = open surface area of leaf

$S_l$  = surface area of leaf ( $m^2$ ) =  $1.10E-02 m^2$  (this study, taken from an average of 20 leaves sampled at random in a temperate environment of various different unknown species)

$w$  = washout rate =  $0.012 \text{ day}^{-1}$  (Chamberlain, 1991)

$S_o$  is assumed to be the number of stomata per surface area multiplied by the stomata surface area.

Number of stomata per vegetation surface area =  $3.00E08 m^{-2}$  (Pachepsky & Acock, 1996).

Stomata surface area is calculated as:

$$S_s = \frac{\pi l_s^2}{4} \quad (9)$$

where:

$l_s$  = width of epidermis  $\approx$  stomatal length (m)

$S_s$  = stomata surface area ( $m^2$ )

$l_s = 2.50E-05 \text{ m}$  (Pachepsky & Acock, 1996). Therefore the  $S_s = 4.00E-10 m^2$ .

## *Deposition of radionuclides*

The deposition of radionuclides is assessed using the simplified approach of a deposition velocity to simulate wet and dry deposition as one lumped parameter.

$$D = C_{air} v_d S_{soil} \quad (10)$$

Where:

$D$  = deposition rate (Bq d<sup>-1</sup>)

$C_{air}$  = activity concentration in air (Bq m<sup>-3</sup>)

$v_d$  = deposition velocity (m d<sup>-1</sup>)

$S_{soil}$  = surface area of soil (m<sup>2</sup>)

The deposition velocity of radionuclides from the atmosphere is different for dry deposition and wet deposition associated with precipitation due to the different processes involved. However, in reality, the processes overlap and a clear separation is often not possible. This is especially true when modelling the deposition of radionuclides over long timescales. In such cases, it should be possible, as is done in common practice, to lump dry and wet deposition and to apply a generic interception fraction for vegetation which is empirically derived from observations (IAEA, 1996). The focus of this study is on annual timescales so a lumped parameter for wet and dry deposition is used which has the advantage of keeping the model simple to use and adapt to other ecosystems. Over sub-annual timescales this approach may become a disadvantage due to the loss of accuracy and the need to have separate dry and wet deposition velocities and to model temporal variations in the rate and type of precipitation.

The data for atmospheric concentration of <sup>137</sup>Cs used in the dynamic model is taken from the fallout data from the BNFL and Sellafield Ltd environmental monitoring reports recorded in the consulting report Bryan et al. (2004). Many of the measurements taken from that report for the Drigg location were unable to record high enough atmospheric <sup>137</sup>Cs concentrations to be detected by the monitoring devices. As a result the values from the Sellafield south-side perimeter recorded in Bryan et al. (2004) were used. The justification is that fallout predominates over Sellafield discharges at all but the nearest distances to the plant, and fallout in the Sellafield locations and Drigg should be similar. Sea-to-land transfer is assumed to be small enough due to the very low sea-to-land transfer at Drigg calculated using the UKAEA model (Bryan et al., 2004) that this is a reasonable approximation.

## *Soil migration of radionuclides*

Concentration Ratios used in the model are calculated from measured soil activity concentrations to a depth of 0.1 m, therefore, this is the depth from which the change in soil activity over time is modelled. This is

modelled using a simple clearance model and assuming a constant deposition rate:

$$\frac{dA_{soil}}{dt} = D - (\lambda + r)A_{soil} \quad (11)$$

where

$D$  = the deposition rate ( $\text{Bq d}^{-1}$ ),

$\lambda$  = the decay constant ( $\text{d}^{-1}$ ),

$r$  = migration rate ( $\text{d}^{-1}$ )

$t$  = time (d) and

$A_{soil}$  = soil activity (Bq)

This has the following solution:

$$A_{soil}(t) = \frac{D}{\lambda + r} + (A_{soil}(0) - \frac{D}{\lambda + r})e^{-(\lambda + r)t} \quad (12)$$

This equation is used to give the migration rate on an annual time-scale by giving values to the parameters and isolating  $r$  for  $^{137}\text{Cs}$ . The time-step used in the model to produce the results section, however, is a monthly time-step so the annual migration rate is divided evenly over the time-steps used in the model runs to produce the results section.

Table 1: Empirical soil data used for calculating radionuclide migration rate of  $^{137}\text{Cs}$  from top 0.1 m of soil.

Source	Location	Date	0.1 m ( $\text{Bq kg}^{-1} \text{ dw}$ )
Rudge (1989)	Dune heath	Jun-86	274
Wood (2010)	Dune heath	Mean (2006)	74.6
		Net loss over 20 yrs	200

Multiplying the activity concentration provided by Rudge (1989) and Wood (2010) in Table 1 by the soil density of  $1250 \text{ kg m}^{-3}$  given by Wood et al. (2009a) and the soil depth of 0.1 m will give the soil activity per unit surface area ( $\text{Bq m}^{-2}$ ). This parameter,  $\zeta_{soil}$ , can be substituted for activity into equation 12 to give the migration rate of activity from the top 0.1 m of soil. Doing this requires a deposition rate per unit surface area,  $\zeta$ , to be substituted in place of  $D$ . Therefore the units for deposition rate in this case are given as ( $\text{Bq m}^{-2} \text{ d}^{-1}$ ), an average over the 20 years from 1986 to 2006 provided by Dr. Mike Wood. Therefore the values used are:

$\zeta = 1.34\text{E-}01 \text{ (Bq m}^{-2} \text{ d}^{-1}\text{)}$  (Dr. Mike Wood pers. corres.)

$t = 7.31\text{E}03 \text{ (d)}$

$\lambda = 6.29\text{E-}05 \text{ (d}^{-1}\text{)}$

$$\zeta_{soil}(0) = 3.43E04 \text{ (Bq m}^{-2}\text{)}$$

$$\zeta_{soil}(t) = 9.33E03 \text{ (Bq m}^{-2}\text{)}$$

Putting these values into equation (12) gives:

$$9.33E03 = \frac{1.34E-01}{6.29E-05+r} + \left(3.43E04 - \frac{1.34E-01}{6.29E-05+r}\right) e^{-(6.29E-05+r)7.31E03}$$

This gives a value of  $r = 1.23E-04 \text{ day}^{-1}$ . However, the data from Rudge (1989) is taken from dunes nearer in proximity to Sellafield and the Low Level Waste Repository than the data from Wood (2010). Therefore, this migration rate can only be taken as a useful approximation for this study.

### *Dose to biota*

The general approach used in this study for developing reference biota follows that recommended by the ICRP 108 publication (ICRP, 2009). ICRP (2009) does include some parameters for reference biota such as shape dimensions. However, these do not have to be used for all reference biota developed for estimating the radiological risk to biota. The reference biota developed for the dynamic model for this study is done using parameter values from different sources besides just ICRP (2009). The parameters used for the reference biota and their sources are presented in Table 4.

Dose is calculated using the approach in Copplestone et al., (2001). The dose to biota is derived primarily through the use of dose per unit concentration (DPUC) factors. These relate the concentration of activity either in the soil or within the organisms to the amount of energy absorbed by the biota. The DPUCs for the model are taken from Copplestone et al. (2001) except for  $^{137}\text{Cs}$  deposited onto the external vegetation. The DPUC values for externally deposited  $^{137}\text{Cs}$  are calculated by this study. For the assessment of dose it is necessary to estimate the fraction of energy which is absorbed by the organism. The dose rates can then be calculated. Therefore, the shape of the organism will have an important influence on the energy absorbed by the organism. In Copplestone et al. (2001) organisms are assumed to have a simplified ellipsoid shape.

The organism dimensions will affect the amount of energy absorbed from ionising radiation. It is important the DPUC values are calculated assuming similar dimensions to the reference organism developed. This means that although a woodlouse is a detritivore it is used to represent the carnivorous beetle as it is the organism used in Copplestone et al. (2001) with the most similar dimensions to the reference carnivore developed for this study. For the assessment of external dose simple formulae are used to estimate dose in an infinite or semi-infinite absorbing medium. The organism mass influences the organism internal activity concentration and so will influence the absorbed dose.

The amount of dose received depends on a number of factors including the radiation emitted by the source, the density of the surrounding media, and the size of the organism. The basic quantity for measuring absorbed energy is the Gray (Gy). This measures the amount of energy absorbed per unit mass of an organism and is usually summed over a specific time period such as an hour to give the units (Gy h<sup>-1</sup>)/(Bq kg<sup>-1</sup>) for the DPUC and (Gy h<sup>-1</sup>) for the dose to biota. Different types of radiation can vary in their biological effects when absorbed so there is the need for an effective dose unit. The energy absorbed is often converted using a weighting factor that varies according to the type of radiation. Dose is calculated as:

$$Ds = C^{biota} \times DPUC^{int} + (f_{soil} + 0.5f_{surface} + f_{air} \times (reduction\ factor)_{radiation\ type})C^{soil} \times DPUC^{ext} \quad (13)$$

$Ds$  = dose to biota (Gy h<sup>-1</sup>)

$C^{biota}$  = activity concentration of biota (Bq kg<sup>-1</sup>)

$C^{soil}$  = activity concentration of soil (Bq kg<sup>-1</sup>)

$DPUC^{int}$  = Dose per unit concentration for internal activity (Gy h<sup>-1</sup>)/(Bq kg<sup>-1</sup>)

$DPUC^{ext}$  = Dose per unit concentration for activity in soil (Gy h<sup>-1</sup>)/(Bq kg<sup>-1</sup>)

$f_{soil}$  = fraction of time organism spends in soil

$f_{surface}$  = fraction of time organism spends on soil surface

$f_{air}$  = fraction of time organism spends above the ground surface

*reduction factor* = factor by which the radiation dose rate above the ground surface is lower than that within the soil itself. This is zero for  $\alpha$  and low energy  $\beta$  radiation and 0.25 for high energy  $\beta$  and  $\gamma$  ray photons.

Here, a new approach is given that allows the dose from radionuclides deposited on to the vegetation surface to be taken into account. The total unweighted dose from surface-deposited radionuclides is then calculated as:

$$Ds_{surf} = \sum_{i=1}^4 C_i^{surf} DPUC_i^{surf} = \frac{SV^{-1}}{\rho_{plant}} \sum_{i=1}^4 \zeta_i^{surf} DPUC_i^{surf} \quad (14)$$

Where  $C_i^{surf}$  is the activity concentration on plant surface per unit mass of plant, equivalent to the activity per unit surface area  $\zeta_i^{surf}$  multiplied by the surface area/volume ratio  $SV^{-1}$  and divided by the plant density  $\rho_{plant}$ .

Furthermore,  $i$  is the summation index for the radionuclides (5 for the free fraction and 4 for the unattached and attached fractions). The Environment Agency R&D 128 random sampling (Monte Carlo) VB code that is used for internal dosimetry was used to provide a summation of the probability of absorption over trajectories selected by choosing at random a point of origin and a point of destination, both inside the ellipsoid representing the grass. This code was used but with modified parameters to establish the point of origin on the ellipsoid surface and modified absorbed energy functions to take into account that, at very low energies, the absorbed fraction for deposited activity tends to 0.5 instead of unity. This approach was used to generate DPUCs for <sup>137</sup>Cs deposited on the vegetation surface (Table 3).

Table 2: DPUCs used in model taken from Copplestone et al. 2001.

<sup>137</sup> Cs	Vegetation	Herbivore	Carnivore	Detritivore	Omnivore
	(Gy h <sup>-1</sup> /Bq kg <sup>-1</sup> )				
Internal, low energy beta	2.4E-07	2.4E-07	2.4E-07	2.4E-07	2.4E-07
Internal, beta and photon	9.5E-05	1.4E-04	1.2E-04	1.3E-04	1.5E-04
<b>Total</b>	<b>9.6E-05</b>	<b>1.4E-04</b>	<b>1.2E-04</b>	<b>1.3E-04</b>	<b>1.5E-04</b>
External, low energy beta	1.06E-10	1.32E-11	7.25E-11	2.64E-11	9.14E-12
External, beta and photon	3.73E-04	3.31E-04	3.51E-04	3.39E-04	3.22E-04
<b>Total</b>	<b>3.7E-04</b>	<b>3.3E-04</b>	<b>3.5E-04</b>	<b>3.4E-04</b>	<b>3.20E-04</b>

Table 3: DPUCs calculated for this study for <sup>137</sup>Cs deposited on vegetation surface

	DPUC for <sup>137</sup> Cs on vegetation surface (Gy h <sup>-1</sup> /Bq kg <sup>-1</sup> )
Internal, low energy beta	1.20E-07
Internal, beta and photon	2.70E-06
<b>Total</b>	<b>2.82E-06</b>

### Reference biota

Default biota parameters for mass and dimensions are taken from the Reference Animals and Plants Report (ICRP, 2009). The shape of biota for assessing dosimetry is simplified by assuming biota take the form of solid ellipsoids. Occupancy factors are taken from Copplestone et al. (2001) while the Concentration Ratios are shown in Table 4. The biohalf-life or  $T_{B1/2}$  for vegetation is taken from observational results from Avery (1996). For the animal biota the data for biohalf-life is calculated using the principle of allometry. This principle is based on experimental data that shows metabolic processes are related to their mass and can be modelled according to the relationship  $Y = \alpha X^\beta$  where  $Y$  and  $X$  are size related measures, and  $\alpha$  and  $\beta$  are constants determined by observational data (Higley et al. 2003). The turnover of radionuclides is thought to

be controlled by metabolic processes and so the allometric function  $T_{B1/2} = 13.22M^{0.237}$  can be used to predict the biological half-life (Higley et al. 2003). The scaling constants 13.22, which includes appropriate unit conversions, and 0.237, which is the allometric scaling factor which relates body mass,  $M$ , to biological elimination of contaminant, for Cs have been taken from parameters used for the FASTer dynamic model in FASSET (2003) which derived the constants from the EPIC report (Beresford et al., 2003).

Reference biota are selected to provide a range of feeding strategies as feeding ecology of animals is important for the transfer of radionuclides between trophic levels and radionuclide uptake (Wood et al., 2009b). The reference biota modelled is selected to provide a range of behaviours and physiologies to allow the results to be extrapolated to a range of biota. The reference biota modelled and the parameters used are shown in Table 4.

Table 4: Reference biota modelled and parameters used.

RAP	Mass (kg)	$T_{1/2\text{bio}}$ (days)	Concentration Ratio (Wood et al. 2009b except vegi spike)	Dimension (cm)			Occupancy factors		
				x	y	z	In Soil	Soil Surface	Air
Earthworm (Detritivore)	5.24E-03	3.81	2.35E-02	10	1	1	1	0	0
Bee (Herbivore)	5.89E-04	2.27	6.53E-02	2	0.75	0.75	0	0.1	0.9
Beetle (Carnivore)	5.89E-04	2.27	1.77E-02	2	0.75	0.75	0	1	0
Rat (Omnivore)	0.314	10.05	1.71E-02	20	7.5	7.5	0.6	0.4	0
Vegetation (spike)	9.40E-04	2.53	6.93E-01	1.30 E-01	5.50E-05	2.50E-04	0	1	0

## **Model scenarios**

### ***Ecosystem selection - Drigg dunes***

The ecosystem being modelled is the heath sand dune part of the Drigg dune ecosystem, a primarily acidic dune system supporting large areas of Atlantic decalcified fixed dunes (Wood et al., 2008). The ecosystem covers about 6 km<sup>2</sup> of land and is bound by the Irish Sea and the inner Esk Estuary. The ecosystem includes embryonic, shifting white, fixed grey, and fixed heath dunes with the majority of dunes consisting of fixed grey and heath dunes (Wood et al., 2008). The dunes are kept in a semi-natural state by grazing livestock. The majority of vegetation consists of marram grass (*Ammophila arenaria*) and red fescue (*Festuca rubra*) (Wood et al., 2008). A varied amount of biota is present with heather and lichens prevalent in varying degrees throughout the ecosystem with protected species such as rare small adder's tongue fern (*Ophioglossum azoricum*) also present. A range of fauna is present including the invertebrate northern dune tiger beetle (*Cicindela hybrida*), birds such as the large skylark (*Alauda arvensis*) and meadow pipits (*Anthus pratensis*), and small mammals including the field mouse (*Apodemus sylvaticus*) and field vole (*Microtus agrestis*). Reptile species include the European adder (*Vipera berus*) and the common lizard (*Lacerta vivipara*) (Wood et al., 2008).

### ***Warm-up run: 1990 – 1995.***

The ICRP Recommendations (ICRP, 2007) recognise three types of exposure situations. Planned exposure is the planned introduction of radioactive sources; existing exposure situations of historical radiation sources; and emergency exposure situations with the unplanned release of radionuclides. The simulating of biota uptake as a warm-up period is necessary as all compartments are empty when the model is initially set up. Having a warm-up period allows the model to reproduce the activity prevailing in the biota before the hypothetical emergency exposure scenario. The model is run using atmospheric concentration data of <sup>137</sup>Cs from 1990-1995 to allow sufficient time for the biota activity compartments to 'warm-up' and allow the assessment of model output for a scenario with low levels of atmospheric <sup>137</sup>Cs. This is done to allow a comparison of model output for low levels of atmospheric <sup>137</sup>Cs with high levels of atmospheric <sup>137</sup>Cs which may occur in an emergency scenario. Appendix 1 gives the empirical data of atmospheric levels of <sup>137</sup>Cs from 1990-1995 which are used in the model from the years 1990-1995. The soil activity concentration is calculated using the empirical data for 1986 taken from Rudge (1989) and the simple clearance model described above.

### ***Emergency exposure scenario: 1996 – 2000.***

A second hypothetical scenario is run to simulate the model results from a hypothetical accidental release of <sup>137</sup>Cs. This is modelled using the empirical deposition data recorded at the Drigg dunes from the Chernobyl incident. This allows the simulation of an emergency exposure scenario with an unplanned, relatively high release of <sup>137</sup>Cs into the atmosphere. The soil activity concentration is calculated using the empirical data for

1986 taken from Rudge (1989) and the simple clearance model described above. The model is run from the years 1996 – 2000, using the deposition data from Bryan et al. (2004). This means there is a switch in the type of empirical data used for the model runs when compared with the warm-up run. The emergency exposure scenario uses the empirical data for the amount of  $^{137}\text{Cs}$  deposited to soil following the Chernobyl incident as measured by Bryan et al. (2004) (specific data cannot be shown in this study as it is confidential due to the report being used for commercial purposes and permission has not been given by the authors to publish the data, only to use in model runs). This is different to the approach used for the warm-up, which has empirical measurements of atmospheric concentration of  $^{137}\text{Cs}$  as the input data and uses a deposition velocity to calculate the subsequent levels of  $^{137}\text{Cs}$  deposited to the soil. The reason for this is a deposition velocity could not be attained for  $^{137}\text{Cs}$  under emergency conditions. The use of the deposition velocity in this study is for long-term low levels of atmospheric  $^{137}\text{Cs}$  and so its use in an emergency scenario will greatly over-predict the levels of  $^{137}\text{Cs}$  deposited to the soil. The main purpose of this study is to investigate the accuracy of the biota uptake and release model developed here and so it is more important to ensure the validity of the environmental data used for the model scenarios. Using the deposition data from Bryan et al. (2004) ensures the hypothetical scenario developed for this study is based on a feasible situation given what is known about the operational past of nuclear installations. As with all other data, the deposition values are averaged over 12 monthly time-steps.

### *Sensitivity analysis*

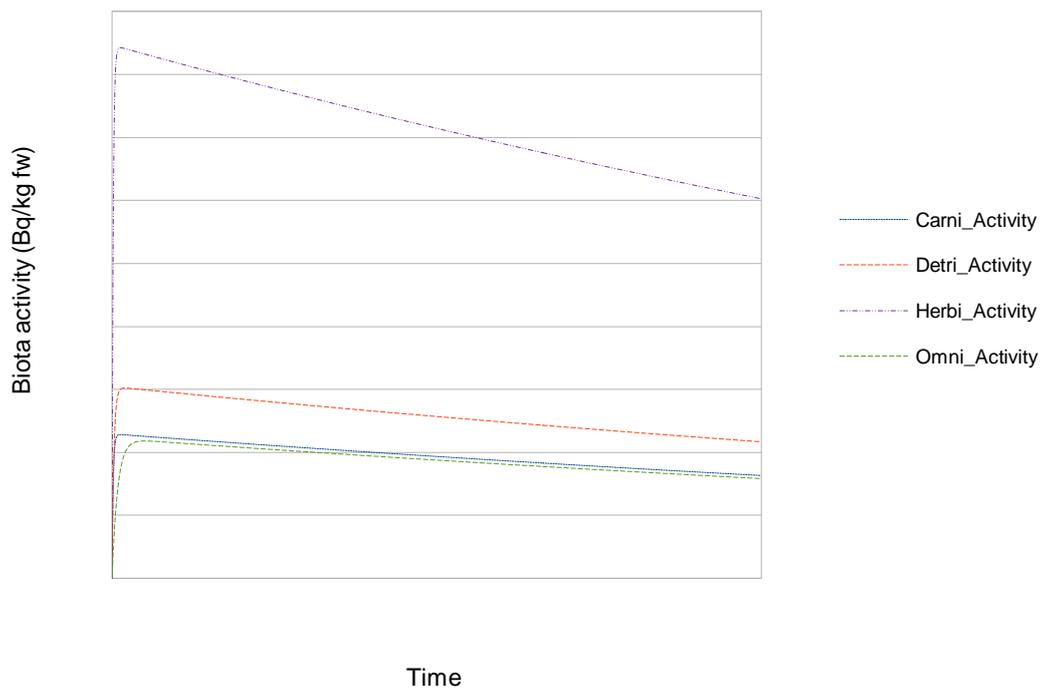
A sensitivity analysis is carried out on the model to investigate the effect of varying a number of different parameters on model output. The effect of varying the Concentration Ratio, biohalf-life, and soil migration rate on the reference herbivore activity concentration during the model warm-up scenario is investigated. The sensitivity analysis varies the parameter values at 1.25, 1.5, and 2 times the original values with results expressed as a % variation from the model output using the original parameter values. This means the sensitivity analysis is the outcome for model output caused by varying the parameter values with a maximum ratio of 2:1. The herbivore results are taken as representative of the other animal biota. The effect of varying the weathering rate for externally deposited  $^{137}\text{Cs}$  on the overall concentration of  $^{137}\text{Cs}$  in the reference vegetation is investigated during the emergency exposure scenario. This is done by varying the weathering rate parameter by 1.25, 1.5, and 2 times the original values. These results are also expressed as a % variation from the model output using the original parameter values.

# Results

## *Warm-up model run*

### *Biota activity concentration*

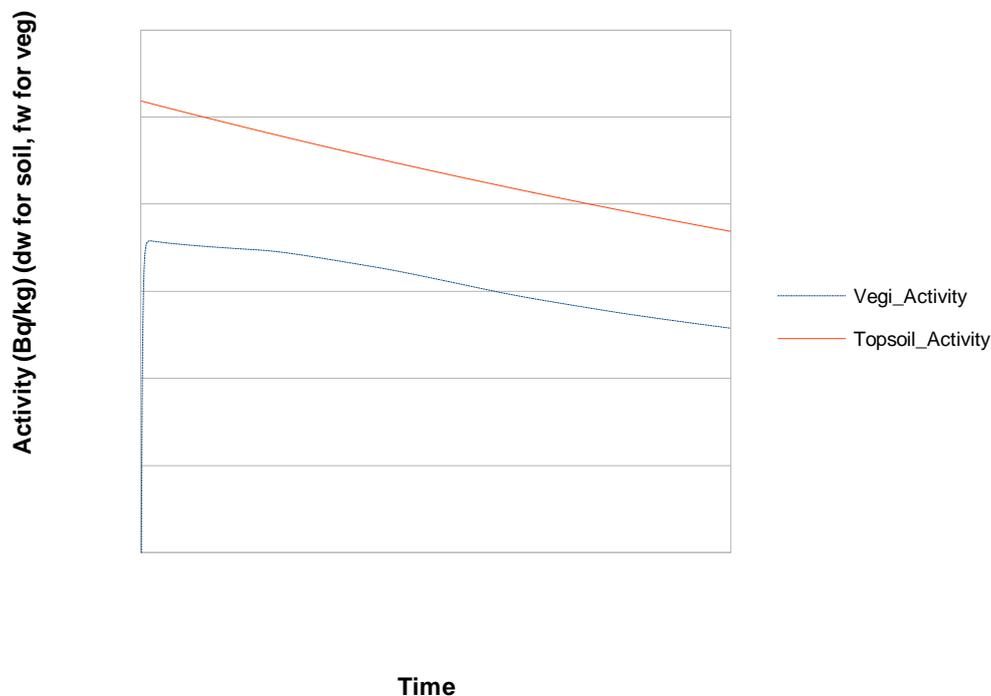
From the reference animals, the herbivore has the highest activity concentration (Fig. 5). Unsurprisingly, the hierarchy of biota activity follows the ranking of Concentration Ratios. It can be seen that the biota reach a peak in activity concentration quickly relative to the annual time-scale with a subsequent steady predictable decrease in activity. The pattern of steady decrease in biota activity (Fig. 5) mirrors the pattern of steady decrease in soil activity due to the soil migration of  $^{137}\text{Cs}$  (Fig. 6). The herbivore activity concentration reaches a peak of  $16.85 \text{ Bq kg}^{-1}$  while the omnivore has the lowest activity concentration at  $4.35 \text{ Bq kg}^{-1}$ . The omnivore peak is delayed slightly compared to the other animal biota while the peak activity of the detritivore is slightly later than that of the herbivore and carnivore. This is expected as the biohalf-life of the reference omnivore is over 4 times that of the reference carnivore and herbivore.



*Figure 5: Predicted biota activity levels for the period 1990 – 1995 with one month time-step. Animal biota activity concentration hierarchy: herbi>detri>carni>omni.*

Vegetation activity concentration is far higher than for animal biota (Fig. 6). The vegetation has a high activity concentration level from the first time step due to the inclusion of the external component and the  $^{137}\text{Cs}$  deposited from the atmosphere. The highest level reached over the 5 years is  $179 \text{ Bq kg}^{-1}$  with the

activity concentration more closely matching soil activity concentration as  $^{137}\text{Cs}$  is removed from the external vegetation. Fig. 6 shows a peak in activity concentration of vegetation very soon after the initial uptake at the start of the model run. An increase in deposition soon after the start of 1991 means there is slight increase in the vegetation activity concentration followed by steady decrease as the deposited  $^{137}\text{Cs}$  are removed from the external compartment. As this happens, the vegetation concentration levels more closely match the trend in topsoil activity concentration. The peak activity concentration is around 10 times that of the nearest animal biota. By the end of the warm-up period the vegetation activity concentration is still around 10 times that of the nearest animal biota. The vegetation activity concentration follows a similar pattern to that shown in Fig. 5 with the overall trend following the decreasing soil activity concentration. However, Fig. 6 does show a slight increase, or 'blip' in the vegetation activity between the years 1991 and 1992. This is due to slight increase in atmospheric levels of  $^{137}\text{Cs}$  recorded at this time and the associated increase in  $^{137}\text{Cs}$  deposited onto vegetation.



*Figure 6: Specific activities for topsoil (0 – 0.1 m) and vegetation from 1990-1995 with monthly time-step. Fresh weight for vegetation and dry weight for soil.*

The impact of including an external compartment can be examined by running the model using just uptake from the soil (Figure 7). This graph shows an almost identical pattern in the activity concentration of vegetation to Fig. 6. The only noticeable exception is there is no slight increase in the vegetation activity concentration between the years 1991 and 1992 in Figure 7. The highest activity concentration reached in Fig. 7 is  $179 \text{ Bq kg}^{-1}$  very quickly after the initial contamination. The overall trend is the same as that followed by the uptake and release of  $^{137}\text{Cs}$  by the animal biota. This means that that the vegetation activity

concentration in this scenario mirrors the trend followed by the soil activity concentration with a steady decrease caused by the soil migration rate of  $^{137}\text{Cs}$ . At the end of the warm-up period the vegetation activity concentration is  $128 \text{ Bq kg}^{-1}$  which is the same as at the end of the model run in Fig. 6 with the activity concentration still much larger than the reference animal biota. The herbivore has the highest animal activity concentration and by the end of the warm-up run this has a concentration level of  $12 \text{ Bq kg}^{-1}$  and so the vegetation has an activity concentration 10 times that of the herbivore even without including the external compartment. This shows that with low amounts of  $^{137}\text{Cs}$  deposition over long time periods the uptake of  $^{137}\text{Cs}$  into the internal compartment dominates the vegetation activity concentration.

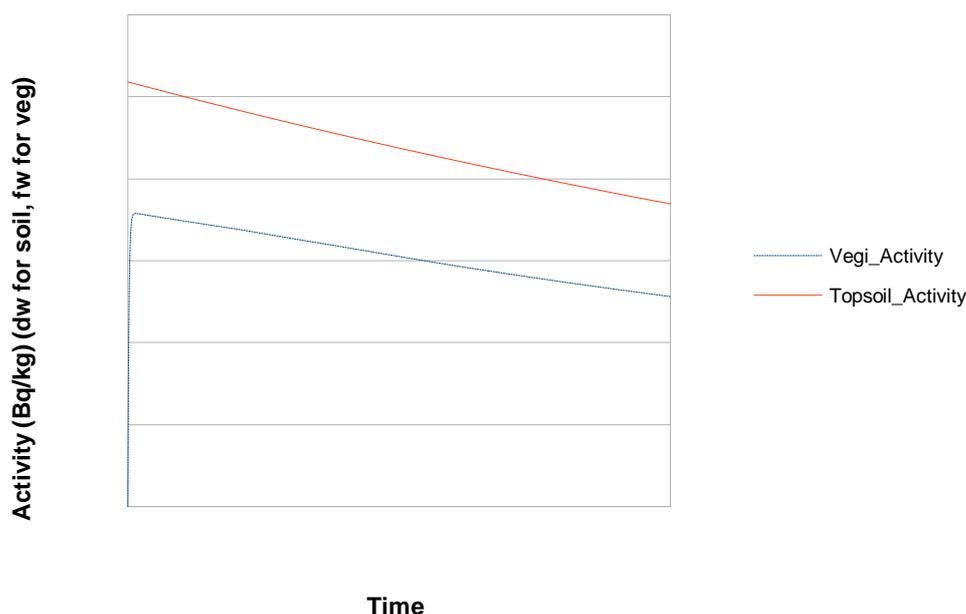


Figure 7: Comparison of vegetation and soil activity concentration with only an internal vegetation compartment from 1990 - 1995 with monthly time-step.

## Dose to biota

The dose to biota over the warm-up period is shown in Figure 8. In assessing the dose to biota all pathways described in the methodology section are included in the assessment. This includes  $^{137}\text{Cs}$  deposited to the external vegetation compartment,  $^{137}\text{Cs}$  deposited and incorporated within the soil, and  $^{137}\text{Cs}$  taken up by biota. The highest dose is received by the detritivore worm while the lowest is received by the herbivore bee. The dose to biota decreases in a predictable pattern that reflects the decreasing soil  $^{137}\text{Cs}$  concentration and the decreasing availability of  $^{137}\text{Cs}$  for biota uptake due to various soil processes such as the migration of  $^{137}\text{Cs}$  to deeper soil. The dose hierarchy follows a pattern of: Detri > Vegi > Omni > Carni > Herbi (Fig. 8). The herbivore has over double the internal activity concentration of  $^{137}\text{Cs}$  compared to other animal biota (Fig. 8) but receives the lowest dose for the warm-up period. This is because of the influence on the total

absorbed dose from external  $^{137}\text{Cs}$  within the environment and other factors besides the internal biota activity concentration of  $^{137}\text{Cs}$ . Life history, particularly the proportion of time spent within the soil, and other parameters such as size dimensions affect the dose that biota receive. The reference detritivore and omnivore are assumed to spend the largest amount of time living in the soil with the detritivore worm assumed to spend its full life within the soil and the omnivore rat spending 60% of the time there.

The highest dose received by vegetation is  $0.08 \mu\text{Gy hr}^{-1}$  and a dose of  $0.06 \mu\text{Gy hr}^{-1}$  by the end of the simulation period. The highest dose received by the detritivore was  $0.09 \mu\text{Gy hr}^{-1}$  and  $0.06 \mu\text{Gy hr}^{-1}$  by 1995. The omnivore and carnivore had highest doses of  $0.07$  and  $0.05 \mu\text{Gy hr}^{-1}$  respectively and  $0.05$  and  $0.03 \mu\text{Gy hr}^{-1}$  by the end of the model run. For herbivore, the highest dose was  $0.03 \mu\text{Gy hr}^{-1}$  and the lowest dose was just under  $0.02 \mu\text{Gy hr}^{-1}$ . Therefore, the reference detritivore had the largest change in absorbed dose over the simulation period while the herbivore had a change of just  $0.01 \mu\text{Gy hr}^{-1}$ .

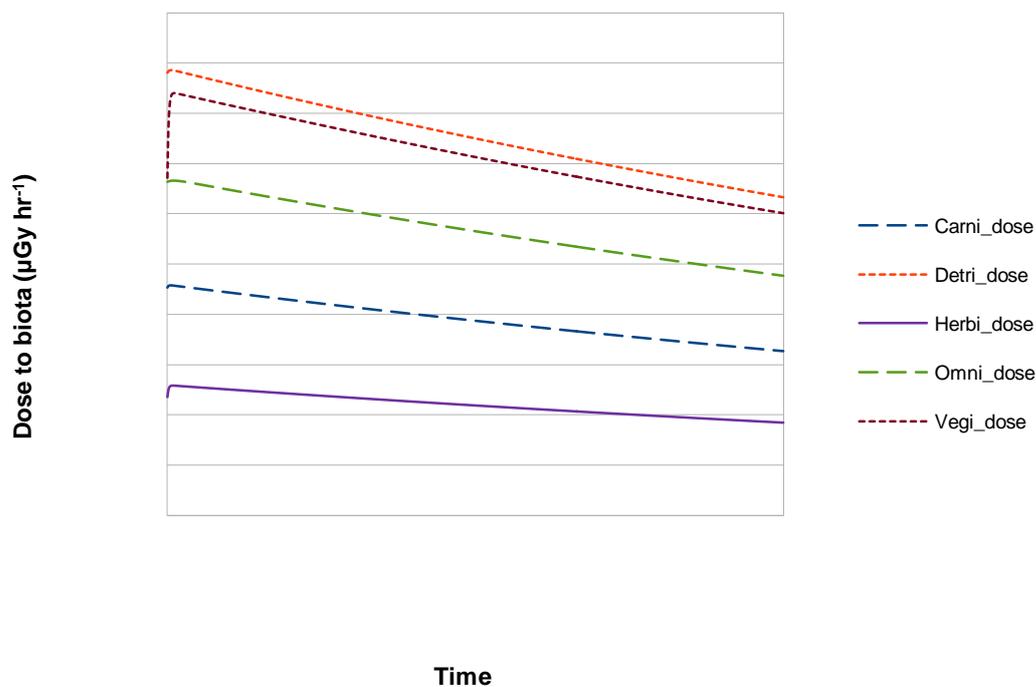


Figure 8: Dose rate of biota for simulation from 1990-1995 with monthly time-step. Dose rate hierarchy is: Detri > Vegi > Omni > Carni > Herbi.

## Model sensitivity analysis

Comparing biota activity concentration with the soil activity concentration shows that, after the initial uptake phase, the activity concentration of biota follow the pattern of soil activity concentration. As soil activity is largely controlled by the migration rate it is important to look at the effect this has on biota uptake. Fig. 9 shows the effect of varying the soil migration rate up to double the rate derived from empirical observation and used as the default value in the model. Increasing the migration rate by 25% shows around a 5% decrease in the activity uptake by the herbivore by the end of the model run. Soil migration has less of an effect closer to the beginning of the model run and so shouldn't have much of an effect during the initial uptake period. However, over time the effect of increasing the soil migration rate has a larger impact on biota activity (Fig. 9).

Varying the Concentration Ratio has a linear impact on the model output for biota activity (Fig. 10). This is similar to previous studies on dynamic modelling (Vives i Batlle et al., 2008). The biohalf-life has an important effect during the initial uptake by biota but has decreasing influence as time increases after the initial uptake (Fig. 11). The effect of varying the biohalf-life is more variable than the other parameters included in the sensitivity analysis. Increasing the biohalf-life by 25% results in varying the herbivore activity concentration by up-to just under 15%. An increase of 50% results in a variation of 25%. A 75% increase in the biohalf-life causes a decrease in activity concentration of up-to just under 35%. Doubling the biohalf-life results in a decrease of the model output of just over 40% during the initial uptake by biota. Therefore, the model is most sensitive to the various factors tested in the order: Concentration Ratio>biohalf-life>soil migration rate.

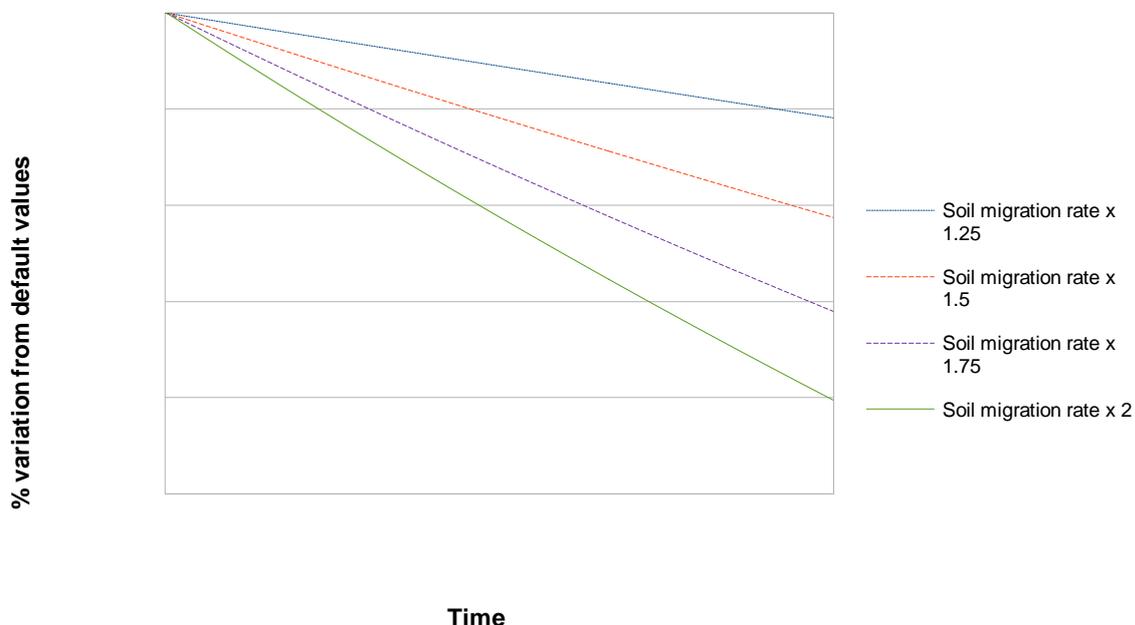


Figure 9: Sensitivity analysis of the effect of varying soil migration rate on herbivore activity concentration with the effect expressed as % variation from their default values. Monthly time-step used.

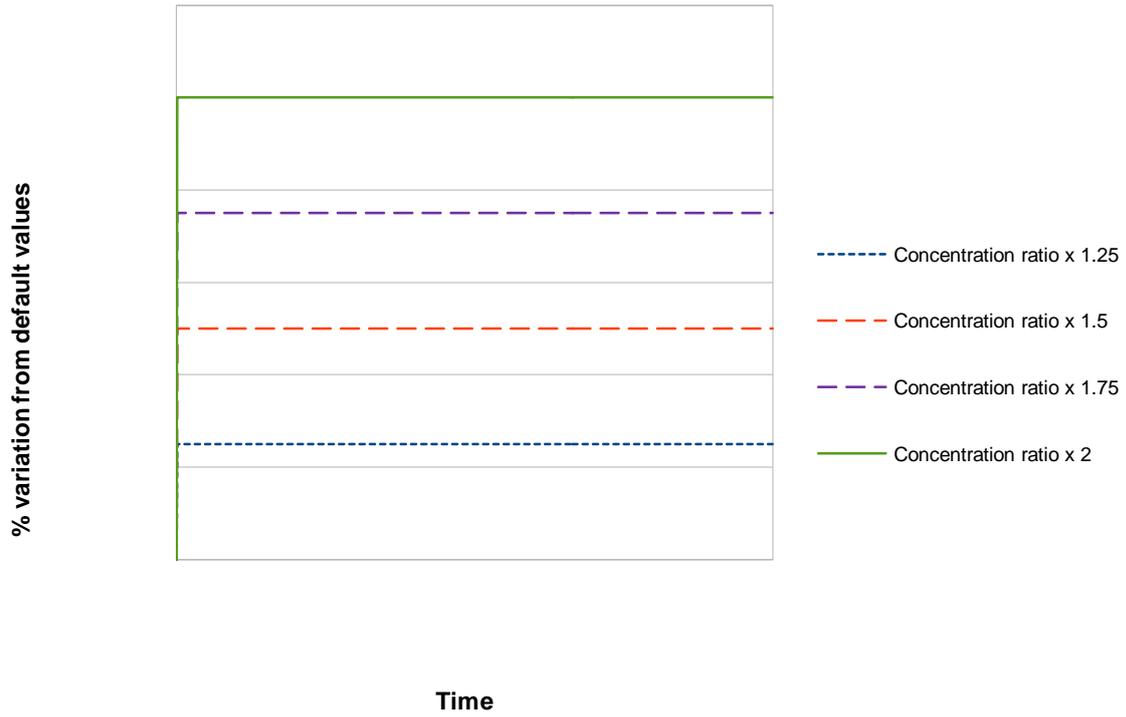


Figure 10: Sensitivity analysis of the effect of varying the concentration ratio on herbivore activity concentration with the effect expressed as % variation from their default values. Monthly time-step used.

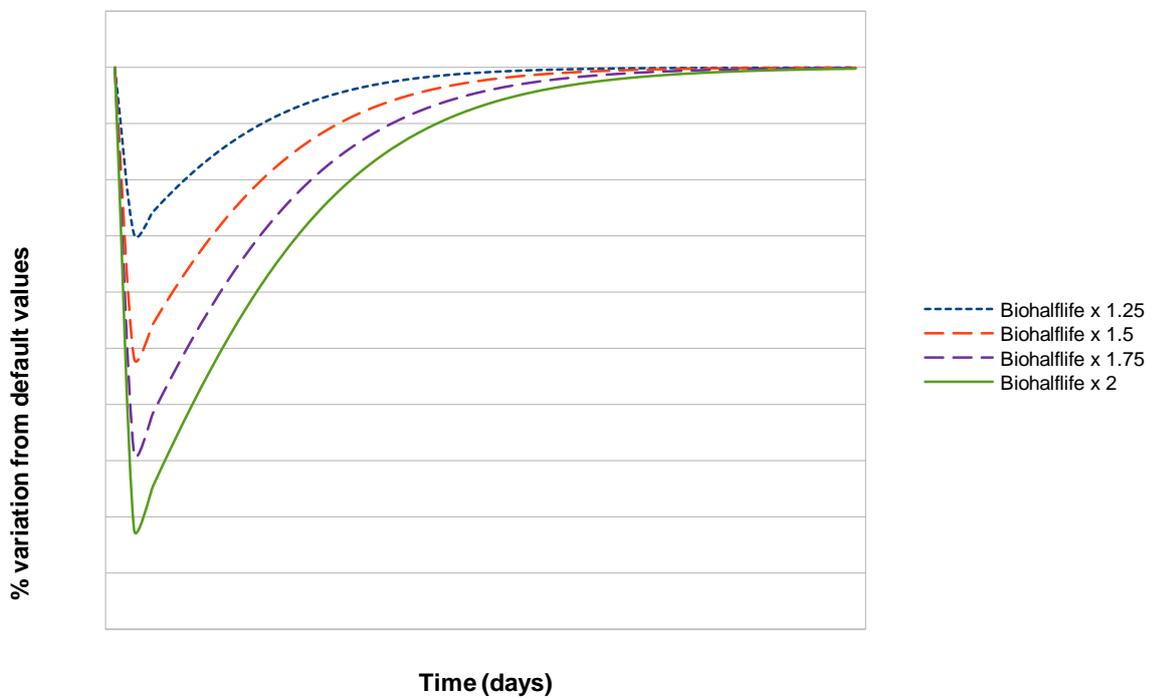


Figure 11: Sensitivity analysis of the effect of varying the biohalflife on herbivore activity concentration with the effect expressed as % variation from their default values. Daily time-step used.

## Accident scenario

Another hypothetical scenario is run to simulate the model results from a hypothetical accidental release of  $^{137}\text{Cs}$ . The hypothetical scenario developed for this study is based on a feasible situation that could arise given what is known about the operational past of nuclear installations. This is modelled using the empirical deposition data recorded at the Drigg dunes from the Chernobyl incident. This allows the environmental impact of an emergency exposure scenario to be assessed with an unplanned, relatively high release of  $^{137}\text{Cs}$  into the atmosphere by using the deposition data from Bryan et al. (2004). Figure 12 shows the model simulated trends from the hypothetical accident scenario simulated from the years 1996-2000. The trends show the vegetation compartment to have a much higher activity concentration than the other compartments. The vegetation compartment reaches a peak of  $3\text{E}04 \text{ Bq kg}^{-1}$  with an activity of  $156 \text{ Bq kg}^{-1}$  reached by the end the of the model run. The peak soil concentration reached is  $1320 \text{ Bq kg}^{-1}$  (dw). The highest activity concentration reached by the animal biota is the herbivore at  $14 \text{ Bq kg}^{-1}$ . The detritivore highest concentration was  $5 \text{ Bq kg}^{-1}$ , carnivore  $3.7 \text{ Bq kg}^{-1}$ , and omnivore  $3.6 \text{ Bq kg}^{-1}$  (Fig. 12).

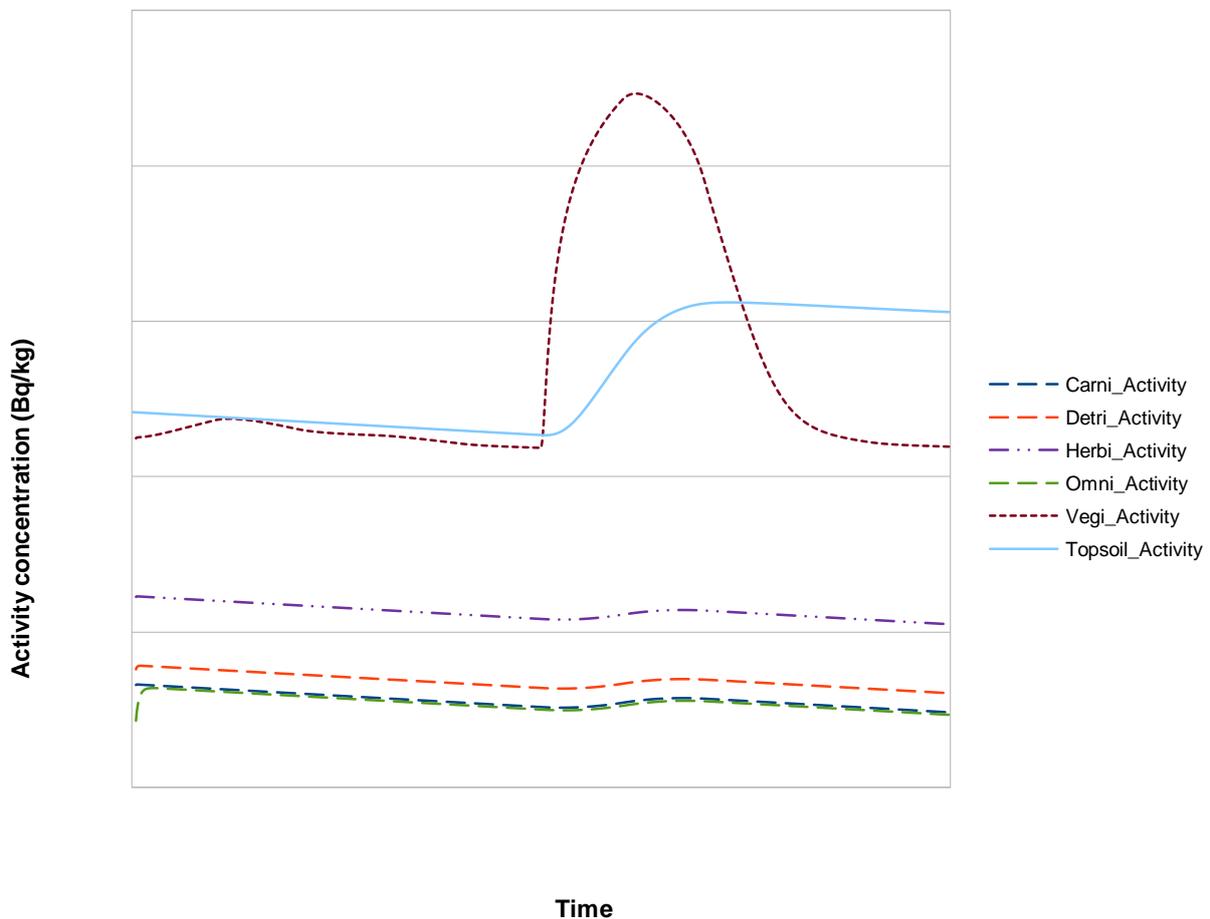


Figure 12: Activity concentration trends (fw for biota, dw for topsoil) from a hypothetical accident scenario using the atmospheric data recorded at Sellafield from  $^{137}\text{Cs}$  released by the Chernobyl accident with a monthly time-step used.

The weathering rate includes all physical processes that contribute to the removal of  $^{137}\text{Cs}$  from the vegetation such as wind removal or being washed off. For  $^{137}\text{Cs}$  radionuclides deposited onto vegetation there is an initial removal of these radionuclides through processes such as bouncing off or being blown off while the remainder are retained on the surface which may be removed through longer term weathering. The weathering rate refers to this longer-term removal of  $^{137}\text{Cs}$  radionuclides as the dose to biota is being assessed on an annual time-scale. The weathering rate used in Fig. 12 is derived from empirical evidence in IAEA (1996) from observations that included woody vegetation. The weathering rate of leafy vegetation that comprises most vegetation at dune ecosystems has been observed to be higher.  $^{137}\text{Cs}$  incorporated into woody plant parts will be less subject to removal by rain and fog than  $^{137}\text{Cs}$  incorporated into leafy plant parts (IAEA, 1996) so the weathering rate used in the dynamic model will give a conservative estimate of  $^{137}\text{Cs}$  removal from the external compartment.

A sensitivity analysis of the weathering rate for the accident scenario shows this parameter can have a big impact on vegetation activity concentration when there is large deposition of  $^{137}\text{Cs}$  (Fig. 13). Doubling the weathering rate decreases the vegetation activity concentration by up to 85%. An increase of the weathering rate by 25% causes a decrease in vegetation activity concentration of up to 45% compared to using the default weathering rate. With the weathering rate at 1.5 times the default value the vegetation activity concentration varies by over 70% while the weathering rate at 1.75 times the default value causes a variation in activity concentration of over 80%. The largest variation in activity concentration doesn't occur at the peak of the vegetation activity concentration but occurs a year later after the peak in vegetation activity concentration. Before the accident event the variation caused by the weathering rate is much less than after there is a significant deposition of  $^{137}\text{Cs}$ . Once there has been a significant period of time of (4 years) after the accident event the weathering rate again doesn't have a large effect on the vegetation activity concentration (Fig. 13). This indicates that only with a high deposition rate will the weathering rate be a significant factor to consider in assessing the activity concentration of vegetation. This is backed up by the very close similarity of results in Figures 6 and 7, as these are scenarios with a low atmospheric deposition rate.

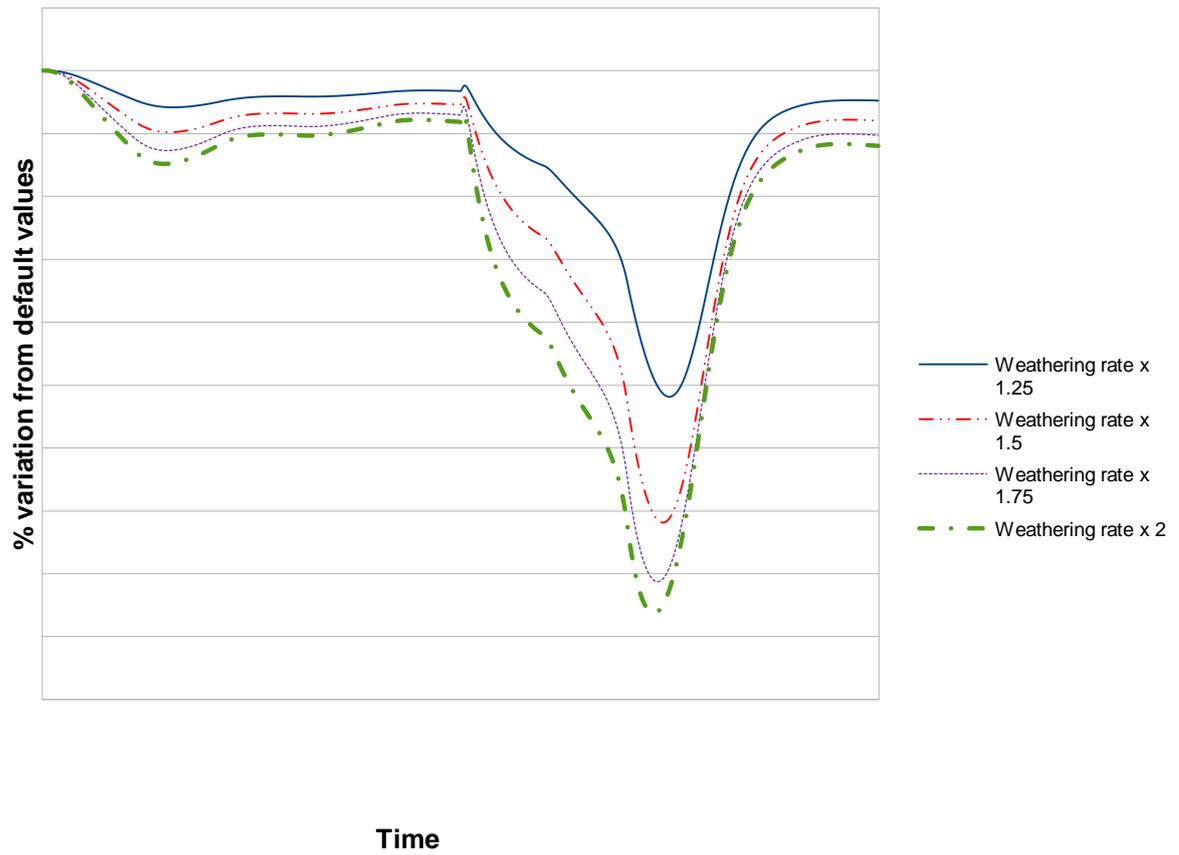


Figure 13: Sensitivity analysis of the effect of varying weathering rate on vegetation activity concentration with effect expressed as % variation from the default weathering rate of 4.22 year<sup>-1</sup>. A monthly time-step is used for model runs.

# Discussion

## *Model simulations*

The hierarchy of biota activity concentrations for the model simulation from 1990 – 1995 can largely be explained in terms of the Concentration Ratio used. The large influence that this parameter has on biota uptake can be seen from the sensitivity analysis of the Concentration Ratio for the herbivore reference animal (Fig. 10). This is consistent with other dynamic modelling approaches (Vives i Batlle et al, 2008) and unsurprising given that all the biota uptake processes are modelled using this lumped parameter. The large activity concentration of vegetation in comparison to the animals during the warm-up scenario is largely due to the conservative Concentration Ratio derived from ERICA in the absence of site specific data.

Biota dose follows a different hierarchy to the activity concentration and so highlights the importance of other parameters used in developing the reference animals and plants. The model uses the ICRP (2009) RAP report values as the default for the animal biota parameters, however, these values can be adapted for specific scenarios. The occupancy factors seem to be more important than the Concentration Ratio for determining the dose rate for animals. This is because of the influence of the external dose from  $^{137}\text{Cs}$  in soil. The detritivore and omnivore are the only two organisms assumed to spend any time in the soil, with detritivore worm assumed to spend its whole life underground and the omnivore rat assumed to spend the majority of time underground. The dose rates to these animals may even be an underestimation as only the  $^{137}\text{Cs}$  from the top 10cm of soil is used in the dosimetry calculations. As they are underground, these animals are more likely than the other biota to be receiving a significant dose from  $^{137}\text{Cs}$  deeper than 10cm. This under prediction may be cancelled out partly by the conservative prediction of  $^{137}\text{Cs}$  being deposited because of the lack of a resuspension factor, although, this is uncertain without more detailed empirical data to validate model predictions. The herbivore bee spends the least time near the soil with 90% of its life assumed to be in the air and this would appear to be largely the reason for having the lowest dose rate for the warm-up period.

The dominance of occupancy factors as a parameter determining biota absorbed dose may be radionuclide specific, which should be considered if the model is expanded to include other radionuclides. The half-life and chemical nature of radionuclides, which determines their interaction with the environment, will affect the importance of different pathways in contributing to the absorbed dose of radiation received by biota.  $^{137}\text{Cs}$  incorporated into the soil matrix contribute much of the dose received by biota from  $^{137}\text{Cs}$  as this radionuclide has a half-life long enough for the  $^{137}\text{Cs}$  radionuclides to become incorporated into the soil matrix in significant quantities. The rate of decay of  $^{137}\text{Cs}$  also means the external dose from the plume released in an accident scenario from a nuclear facility is unlikely to be a significant factor in determining dose to biota. However, for radionuclides with a short half-life such as  $^{131}\text{I}$  the dose from the plume should be considered. Therefore, the dynamic model in this study isn't suitable for considering short lived radionuclides

such as  $^{131}\text{I}$  without including pathways that are important in the short-term after the initial release of radionuclides in an accident scenario. This is likely to include pathways such as the resuspension of radionuclides from the soil, deposition of radionuclides onto animal fur and skin, and the inhalation of radionuclides. The chemical nature of  $^{137}\text{Cs}$  also means that the pathways most significant in determining the absorbed dose of biota will be different for radionuclides with a significantly different chemical nature. For example, for radionuclides that are less available for uptake through the plant rooting system such as  $^{241}\text{Am}$ , the biological pathways are likely to be of less importance while the physical pathways determining the interaction of radionuclides with the soil matrix are likely to be more important. This is particularly the case for long lived radionuclides such as  $^{238}\text{Pu}$ .

The sensitivity analysis of the weathering rate for the accident scenario resulted in the largest variation from the default occurring a year after the peak activity concentration. It is not clear why this should be the case. The effect that varying the weathering rate has with a large deposition of  $^{137}\text{Cs}$  compared with the warm-up scenario shows that the inclusion of a vegetation external compartment is only important for when there is a large deposition rate. An important process for coastal environments not considered as part of the deposition of  $^{137}\text{Cs}$  is the sea-to-land transfer. Hill et al. (2008) indicates this is not as significant a process for the transfer of  $^{137}\text{Cs}$  but sea-to-land transfer is particularly effective for transporting actinides. If the model is expanded to include these radionuclides the influence of sea-to-land transfer should be included.

Fig. 13 shows a difference in shape before and after the peak values of % variation from the default weathering rate. Before the large deposition of  $^{137}\text{Cs}$  that results from the emergency scenario throughout the year 1996 there is more of a fluctuation in vegetation activity concentration caused by uptake and loss processes. After the large deposition of  $^{137}\text{Cs}$  it is the concentration of  $^{137}\text{Cs}$  in the external vegetation compartment that dominates the vegetation activity concentration. Therefore, the smooth decrease in activity concentration that is seen in Fig. 13 after the peak value is as a result of the weathering rate being the dominant process in affecting the activity concentration as the vegetation gradually loses the deposited  $^{137}\text{Cs}$  from various processes such as removal by rainwater, which is modelled by the weathering rate parameter.

The sensitivity analysis for the weathering rate does not produce a linear response in model output. With a large deposition event the weathering rate becomes more influential in determining the vegetation activity concentration. However, from the sensitivity analysis it can be seen that the impact of the weathering rate decreases with each increase of the parameter value. This means increasing the weathering rate does not produce a linear effect on the vegetation activity concentration as the influence of other parameters such as root uptake become a limiting factor on the level of influence the weathering rate can have on model output.

The sensitivity analysis varied parameter values from 1.25-2 times the model default values for equal comparison of the effect each variable has on model output. However, this has a limitation as the possible

range of values for the selected variables may be more than a maximum of double. For example, the range of Concentration Ratio values found in Wood et al. (2009b) for carnivorous invertebrates range by an order of magnitude. The biohalflife of  $^{137}\text{Cs}$  is calculated using an allometric approach and so the range of values for this parameter will be similar to the range of organism sizes found in an ecosystem. For soil migration rate, Kirchner et al. (2009) reports 11 measured values of the migration rate for radiocaesium from Chernobyl fallout with the maximum value around 8.5 times the lowest recorded value. For the weathering rate, Chamberlain (1991) reports a range of empirical values for radionuclides from 0.023 - 0.6 day<sup>-1</sup>. The large range of Concentration Ratio values in comparison to the other variables further highlights the importance of the Concentration Ratio parameter in terms of the large impact it can have on model accuracy.

The results don't consider the impact of biota life cycle or span. A correction factor is used by FASSET (2003) to estimate the activity concentration correcting for lifespan:

$$C_{corr}(t) = C(t) - \frac{C(t - E_{life})}{1 + k} \quad (15)$$

Where:

$C_{corr}(t)$  is the corrected activity concentration in the organism at death (Bq kg<sup>-1</sup>)

$C(t)$  is the activity concentration in the organism calculated without consideration for the life time (Bq kg<sup>-1</sup>)

$k$  is the effective release rate of the radionuclide from the animal body by radioactive disintegration and via biological processes (d<sup>-1</sup>)

$E_{life}$  is the expected life time of the organism (d)

This is useful for comparing the model output with empirical data with regards to model validation. That is outside the scope of the current study where the aim is to reproduce realistic overall trends in biota activity concentration.

## ***Model parameters***

The model can be separated into three elements. The physical component, biological processes, and dosimetry. Each part contributes to the overall model uncertainty. Analysis of the different parameters for each of these elements is given below.

### ***Physical processes***

The physical component consists of deposition, weathering from vegetation, and soil migration of radionuclides. The physical component has a large amount of simplification for each of the processes modelled and so contains a large amount of uncertainty. The deposition of radionuclides is controlled by a number of factors including: particle diameter, physical processes such as Brownian motion and impaction (Garland, 2001), and atmospheric conditions such as wind speed, precipitation, and boundary layer height (Nelson et al., 2002). The representation of these different processes and factors is often done by representing dry deposition and scavenging of radionuclides by precipitation with the use of a simple deposition velocity,  $v_d$  (Sportisse, 2007). More detailed models will represent the physical processes behind particle deposition. For example, the gravitational settling velocity of particles can be modelled mathematically using Stokes formula (Sportisse, 2007).

Radionuclides can be removed by wet deposition through two processes of rainout where particles are impacted by falling raindrops and washout where particles are incorporated into clouds by acting as condensation nuclei. Wet deposition is modelled as a separate process in more complex models such as the NAME pollutant dispersion model developed by the UK Met Office after the nuclear accident at Chernobyl. This includes a scavenging coefficient defined as  $\Lambda = aR^b$  where  $\Lambda$  = scavenging coefficient,  $R$  is the rainfall rate and  $a$  and  $b$  are coefficients based on observational data for different types of precipitation (e.g. dynamic, convective, rain and snow) and the two different deposition processes: rainout and washout (Nelson et al. 2002). In Nelson et al. (2002) the scavenging coefficient is given in units of  $s^{-1}$  and the rainfall rate as  $(mm\ hr^{-1})$ . The approach in this study simplifies these processes into one lumped parameter: the deposition velocity. The main purpose of this study has been to simulate biota uptake and release dynamically. This means less consideration has been given to physical processes. A more complete study would model the deposition of radionuclides and the interaction of radionuclides with soil and loss processes from vegetation in more detail.

The advection and dispersion of radionuclides through the atmosphere is not included in this model as empirical atmospheric concentrations are used. A number of modelling approaches can be used to simulate the atmospheric transport of radionuclides. Many of which are adapted from and used for the application of general pollutant transport. The ADMS model developed by CERC environmental software company is often used in industry for predicting the atmospheric transport of radiological plumes (Carruthers et al., 1994). The spread of a plume and the spread of radionuclides from different emission sources relies on a complex set of parameters and factors such as the variation of the level of turbulence in the boundary layer of the atmosphere. It is outside the scope of this study to investigate the dispersion of radionuclides from their source.

An interception factor can be used to calculate deposition onto vegetation surfaces (Chamberlain, 1970). This models wet and dry deposition together with Chamberlain (1970) providing a formula that relates the interception fraction with the amount of standing biomass:  $\varphi = 1 - e^{-\mu B}$  where  $\varphi$  = interception factor,  $B$  =

above ground biomass (dry weight) of vegetation per unit area ( $\text{kg m}^{-2}$ ),  $\mu$  = interception coefficient ( $\text{m}^2 \text{kg}^{-1}$ ). The interception factor includes wet and dry deposition. Over shorter time periods it is more appropriate to model wet and dry deposition separately but many modelling approaches model both processes using one parameter but have a higher deposition velocity to account for the increased deposition during rainfall (IAEA, 1996).

The dynamic model developed in this study assumes a percentage of deposited  $^{137}\text{Cs}$  is intercepted by the vegetation. This percentage is equal to the percentage of ground covered by the vegetation. The effect of surface roughness on the interception of  $^{137}\text{Cs}$  is not included in this study. Bunzl et al. (1989) measured total deposition of caesium isotopes and  $^{106}\text{Ru}$  in a spruce forest and nearby grassland following the Chernobyl incident. The difference in deposition between the two surface types was equivalent to a deposition velocity of  $5 \text{ mm s}^{-1}$  for  $^{137}\text{Cs}$  and  $6.6 \text{ mm s}^{-1}$  for  $^{106}\text{Ru}$ . Future development of the dynamic should look to improve the accuracy of vegetation interception by including the effect of surface roughness. The large activity concentration of vegetation during the dynamic model accident scenario highlights the impact of including an external vegetation compartment. Assessment tools that don't explicitly model radionuclides deposited onto vegetation surfaces may be significantly underestimating the risk to biota during an accident scenario. This is backed up by empirical evidence that shows root absorption to be the dominant route of Cs uptake by vegetation until 1986 at Drigg. The Chernobyl event meant direct surface deposition became the more important route of Cs accumulation by vegetation (Rudge et al. 1993).

The processes of wet and dry deposition are two separate processes. However, in reality, the processes overlap and a clear separation is often not possible. As the results of Hoffman et al. (1992) indicate, values of vegetation interception remain nearly constant if the plants can dry between the rainfall events. However, such simplified approaches are not appropriate for the modelling of deposition over short time periods. At sub-annual scales the impact of variations in rainfall and the need to model this explicitly becomes important rather than having just one deposition velocity.

A potentially important factor not accounted for is the temporal variation in the rate that determines loss of deposited radionuclides from the external vegetation compartment. Analysis of the removal rate from different crops due to a mixture of dry and wet Pu deposits showed the weathering rate,  $WR$ , to vary from  $WR = 0.025 \text{ d}^{-1}$  to  $WR = 0.065 \text{ d}^{-1}$  (IAEA, 1996). Chamberlain (1991) reports a range of loss rates from vegetation from  $2.3 - 6.9\text{E-}07 \text{ s}^{-1}$  with the rate of loss not critically dependent on the type of radionuclide, the size of the particles or variability in rainfall. Bondietti et al. (1984) measured the time for activity concentration to decrease by half to be 130 d for  $^7\text{Be}$  on semi-dormant pasture. This is the highest value reported by IAEA (1996). This is largely due to the differences in the rate of loss processes such as the shedding of leaves and cell turnover between dormant and growing vegetation. Seasonal factors have been shown to have an effect on vegetation weathering and radiation concentration. With the highest growth rates occurring from spring to autumn this has a dilution effect on activity concentrations (IAEA, 1996). The life stage of vegetation being modelled may also be a factor as this can affect uptake and excretion rates. Growth dilution processes have been observed to slow down as a plant grows (IAEA, 1996).

IAEA (1996) has shown there is a rapid initial phase in the removal of radionuclides deposited on the surface of vegetation followed by another long-term phase of removal over a period of 60 d. In one study, a fine spray of Chernobyl rainwater was applied to grass grown in pots. One half of the pots were protected against rain, but watered during the whole experimental period with the other pots not protected against rainfall. For the uncovered plots the weathering rate is about a factor of 2 shorter than for the plots protected against rainfall. In both cases, the initial loss is very rapid followed by a phase of slower removal of radionuclides from the vegetation surface (Ertel et al. 1989). The dynamic model developed for this study uses a single half-life derived from the slower, long-term loss of radionuclides to describe the weathering of deposited  $^{137}\text{Cs}$ . Given the observed time-dependence of weathering, this could lead to an over-estimation of the contamination of plants after a large deposition event. However, the use of a single weathering rate is appropriate for estimating contamination due to routine releases or for estimating the concentrations over annual timescales. (IAEA, 1996). If the model were to be adapted for sub-annual timescales then it may be necessary to have weathering rates to cover both the rapid initial loss and slower long-term weathering.

The sensitivity analysis shows that soil migration rate has increasing importance on the effect of biota activity concentration as time increases. The default value of  $0.045 \text{ m year}^{-1}$  used in the model is an empirically derived value. It is possible to calculate the theoretical migration rate using rainfall divided by a retardation factor:

$$r = \frac{R}{\delta(\omega + \rho_{soil} K_d)} \quad (16)$$

where:

$r$  = migration rate in soil ( $\text{y}^{-1}$ )

$\delta$  = soil depth (0.1 m)

$\rho_{soil}$  = soil density (loose sand) =  $1442 \text{ kg m}^{-3}$  (Curry et al., 2004)

$\omega$  = Porosity = 0.463 (Curry et al., 2004)

$K_d$  = distribution coefficient for  $^{137}\text{Cs}$  in sand =  $0.27 \text{ m}^3 \text{ kg}^{-1}$  (IAEA, 2010)

$R$  = Average rainfall rate at Drigg =  $1.1 \text{ m y}^{-1}$  (Dr. Andy Smith, pers. communication)

This gives a migration rate of  $0.03 \text{ m year}^{-1}$  which shows that the empirical value for Drigg dunes used in this study is slightly faster than expected. However, this is a close agreement plus the rainfall data for Drigg dunes is an average value and so the theoretical calculation might match the empirical data more accurately if more accurate rainfall data is used.

The above calculation is a simplification of the more sophisticated modelling of soil migration that can be done using the convection-dispersion modelling approach (CDE). The CDE approach assumes that the

convective velocity is equal to the mean pore water velocity with the retardation effect applied. The horizontal migration of radionuclides can be modelled using a similar approach by applying the retardation factor to an effective dispersion coefficient. The approach used in this study is considered a 'black-box' approach and, with the depth of soil compartment being arbitrarily set, the CDE model is considered the preferred approach (Kirchner et al., 2009). However, the close agreement between the simplified theoretical approach and the 'black-box' approach, and as the black-box compartmental approach is based on empirical data from the Drigg dunes, the default migration rate should be accurate over annual time scales for this study.

The basic physical processes controlling radionuclides in soil is the convective transport by water flow, dispersion caused by spatial difference in convection velocities, movement by nuclides within liquid by the process of diffusion, and the physico-chemical interaction with the soil matrix (Kirchner et al., 2009). The common approach for the sorption of dissolved radionuclide ions to the soil matrix is the use of the distribution coefficient:

$$K_d = \frac{C_s}{C_l} \quad (17)$$

where:

$K_d$  = the distribution coefficient ( $L\ kg^{-1}$ )

$C_s$  = activity concentration for the solid phase ( $Bq\ kg^{-1}$ )

$C_l$  = activity concentration for the liquid phase ( $Bq\ L^{-1}$ )

This assumes that the activity concentration in the solid phase is in equilibrium with the activity concentrations in solution and that exchange between these phases is reversible (IAEA, 2010). The time since a radionuclide is incorporated into the soil can affect the  $K_d$  value as a fraction of the radionuclides may become fixed by the solid soil phase (IAEA, 2010). A number of soil factors can affect the sorption of radionuclides such as the cation exchange capacity (CEC) and the radiocaesium interception potential (RIP) (IAEA, 2010). This process of soil fixation is assumed to be accounted for in this study by the conservative approach to estimating the migration of radionuclides from the plant rooting zone.

### *Biological processes*

The main source of uncertainty in predicting the uptake of radionuclides by biota is the Concentration Ratio. Figure 11 shows the biohalf-life ( $T_{B1/2}$ ) parameter can also have a large influence on biota activity concentration in the early stages of the warm-up scenario, when the organism compartment has low levels of activity concentration. The model predicts a much larger  $^{137}Cs$  uptake for vegetation compared to animal biota. The Concentration Ratio used for vegetation is taken from the ERICA database as no site-specific

value was available. The ERICA approach generally provides an over prediction for  $^{137}\text{Cs}$  as this is usually used as a regulatory screening model (Wood et al., 2008). Future model development should derive more accurate Concentration Ratios for vegetation in dune ecosystems. Concentration Ratio values derived from sites more appropriate to the Drigg dunes, i.e. closer in location or from similar ecosystems, may already be available in the literature that haven't been picked up by this study. If such Concentration Ratio values were used in the model these are likely to produce more accurate model output.

Using the Concentration Ratio provides ease of use as just one parameter is required. However, this introduces a larger amount of variability. Other modelling approaches attempt to model radionuclide uptake through food ingestion and inhalation rates (Sample et al., 1997). An allometric approach can be used to simulate uptake and, as used in this study, turnover of radionuclides (Higley et al., 2003). The parameters used were taken from Beresford et al. (2003), which was concerned with predicting the uptake of radionuclides by Arctic biota. This may lead to uncertainty within the model but the parameters from Beresford et al. (2003) were the most appropriate available. The reason for using the Beresford et al. (2003) parameters is because these parameters were derived from biota most similar to those found in the Drigg dune ecosystem which could be found by this study. Parameters more accurate for sand dune biota may already be available that were missed by this study. Further research is required to derive more accurate allometry parameters for dune biota. The use of allometry means the model is assuming the release rate is dependent on animal size. Higley et al. (2003) has shown this to be an accurate way of estimating radionuclide release in biota.

It has been suggested that inhalation may be a secondary route of radiocaesium uptake in terrestrial ecosystems (Avery, 1996; Copplestone et al., 2000). However, due to high measurement uncertainties, it is not possible to determine whether inhalation of  $^{137}\text{Cs}$  is contributing to the tissue distribution observed. The model also doesn't explicitly include dose from radionuclides deposited directly onto fur or skin. Also, the current approach assumes that radionuclides are uniformly distributed throughout the biota. This is a simplification as radionuclides accumulate more in some tissues of the organism than in others (ICRP, 2009). This is difficult to model as there are few consistent datasets that exist for the distribution for of radionuclides in terrestrial biota tissue (ICRP, 2009). This can be particularly important for larger animals higher up in the food chain such as large mammals where it is important to consider the dose to specific organs such as the liver and gonads. An approach that can be used is to modify the dosimetry methodology by modelling the organs as simplified ellipsoids within the body and considering the dose from radionuclides homogeneously distributed within the body and from a radiation source within the organ (ICRP, 2009).

Many sand dune small mammals typically have a home range that is larger in sand dune ecosystems compared to other habitats such as woodland (Akbar and Gorman, 1993) and so organisms may be more likely to migrate out of the area being considered, resulting in a different dose than predicted. Seasonal variations in body burden have been observed in woodland ecosystems (Toal et al., 2001). This has been attributed to seasonal differences in the activity concentration of food that form part of the mammal's dietary

intake. For example, the consumption of fungal fruiting bodies increases autumnal body burden (Toal et al., 2001).

The time-step used in the model is monthly. This has the potential to capture seasonal variations in body burden and the underlying physical and biological processes provided data that captures these variations is used. The data used in the model is annual data and so seasonal variations discussed above aren't captured. The annual data is divided evenly over the 12 monthly time-steps. While using a monthly time-step allows seasonal variations in processes affecting the uptake of radionuclides to be modelled it does mean the model isn't appropriate for some radionuclides. Using the monthly time-step isn't appropriate for radionuclides such as  $^{131}\text{I}$  that may be significant in the inventory of radionuclides released in an emergency scenario. This radionuclide has such a short half-life that a sub-monthly time-step is required to accurately estimate the absorbed dose. This also means that the significant short-term processes important soon after an emergency scenario for  $^{131}\text{I}$  such as the initial shine path and dose from the plume released can't be effectively modelled using the monthly time-step.

The model is generally applicable to a wide range of biota as the biological processes can be adapted for a wide range of organism sizes and habitat ranges due to the use of allometry and occupancy factors. For some biota, it may be important to consider different life stages of an organism. The different dimensions and behaviours exhibited by some biota as they change life stages such as from caterpillar to butterfly or as amphibians develop will significantly affect the absorbed dose received and this must be considered if the model is applied to specific species or extended for other reference biota.

## *Dosimetry*

Various areas of improvement are required for assessing dose to biota. Models for the tissues of vegetation are lacking such as for the xylem and meristem (Pentreath, 2009). This study highlights the need to consider radionuclides deposited externally onto vegetation. Dose assessment may also be improved by considering the non-uniform distribution of internally incorporated radionuclides such as the distribution of radionuclides specifically within internal organs, shielding layers such as skin or fur and using geometries more accurate in shape than simple ellipsoids.

Also there is a requirement for further study into the Relative Biological Effectiveness (RBE) of radiation for non-human biota (Pentreath, 2009). This is particularly true for alpha-emitting radionuclides as models tend to have high variation in their dose assessment from alpha-emitters (Vives i Batlle et al., 2007) and many animals and plants have high levels of naturally occurring alpha-emitters and the use of weighting factors are useful to normalise background dose (Pentreath, 2009). However, dosimetric parameters contribute less to the overall uncertainty than the radioecological parameters (Avila et al., 2004).

Salbu (2009) highlights a need to research the effects of low chronic doses on biota. This study backs this up with the dose to biota in the warm-up scenario coming from low chronic doses. The main concern for biota, unlike human radioprotection, is population level effects. An effect at population level means there is

an adverse effect at the individual level, but an effect on individuals will not always result in population level effects. The endpoints of most significance for environmental protection are those most likely to affect population size and structure: morbidity, mortality, reduced reproductive success, and chromosomal damage.

The lethal dose for 50% of the population  $LD_{50}$  for the wild grass barley has been measured at 20 Gy (Holt and Bottino, 1972). A dose rate of over 24 mGy day<sup>-1</sup> has been observed to reduce the fertility of rye grass (Holt and Bottino, 1972). The dose measured during the model run 1990-95 is well below this value. The highest animal biota dose was measured for the reference detritivore worm which had a peak dose of 0.09  $\mu$ Gy hr<sup>-1</sup>. 5-20 Gy dose has been observed to inhibit the growth of epidermal cells of earthworms (Suzuki and Egami, 1983) and 264 mGy day<sup>-1</sup> to cause abnormalities in offspring earthworms (Hertel-Aas et al., 2007). Again the modelled dose to reference biota is well below this value. For small mammals a dose of 10mGy day<sup>-1</sup> may have some fertility effects with previous studies finding reduced germ cells in male and female rats (Erickson, 1978). Mortality effects are seen at 23 mGy day<sup>-1</sup> with a reduced lifespan of 11-30% seen in mice. Small insects are more resilient to radiation effects with a range of  $LD_{50}$  values for adults observed from 20-3000 Gy (ICRP, 2009). The results for the 1990-95 run would appear to show that there is no significant risk to the environment in this time period. Indeed at very low doses there is evidence that some single-celled organisms benefit from radiation exposure (Planel et al., 1987).

## ***Regulatory modelling***

The need for dynamic modelling for non-human biota in a regulatory context is a topic of debate within industry (Brownless, 2007). Many factors are under consideration and this includes the need to take action for species, even endangered species, during the acute phase of an accidental release, when non-equilibrium conditions are most likely to happen, when there is likely a risk to humans (Coppelstone et al., 2004). Here is presented a way to dynamically model <sup>137</sup>Cs uptake using simple parameters. Other dynamic models have been developed that are based on ingestion rates of food (Toal et al., 2001). Such an approach requires research to find out the dietary contents of biota and how this varies seasonally. The approach by Toal et al. (2001) allows the calculation of seasonal variations in activity concentration required for a sub-annual dynamic approach. Increased <sup>137</sup>Cs concentrations in roe deer have been observed in summer and autumn, attributable to increased ingestion of fungi during these months (IAEA, 2009).

The dynamic model produced by this study may be able to run on a sub-annual basis but this would require the researching of seasonal Concentration Ratio values for which no database currently exists. This may allow the time lag in transfer of <sup>137</sup>Cs between trophic levels observed in earlier studies (Reichle & Crossley, 1969) to be incorporated into the dynamic model. A seasonal fluctuation in vegetation from Drigg in <sup>137</sup>Cs concentration was observed with a second peak of Chernobyl <sup>137</sup>Cs in the late summer or autumn of 1987. Several processes may contribute to this phenomenon, including remobilisation of Cs prior to autumn die-back, leaf ageing and seasonally reduced translocation (Rudge et al., 1993). It is these sorts of sub-annual variations that aren't adequately modelled annually and that would need to be included in a sub-annual modelling approach.

Using the model in a regulatory context also requires considering the applicability and implications of incorporating additional radionuclides that have different properties to  $^{137}\text{Cs}$ . As discussed above, pathways need to be added to for short lived radionuclides such as  $^{131}\text{I}$  to include processes such as the resuspension of radionuclides from the soil, deposition of radionuclides onto animal fur and skin, and the inhalation of radionuclides. These pathways will be more important in assessing the absorbed dose in the short-term after the release of radionuclides. Also, the monthly time-step used in the model will not be appropriate for radionuclides with a half-life on a sub-monthly timescale as this will not adequately capture the absorbed dose received by biota. The chemical nature of  $^{137}\text{Cs}$  means it is more available for biota uptake relative to many other radionuclides. This means for radionuclides such as  $^{241}\text{Am}$ , the biological pathways are likely to be of less importance while the physical pathways such as sea-to-land transfer will be more important in determining dose to biota. The interaction with the soil matrix will be particularly important for longer lived radionuclides that aren't as readily incorporated into the food-chain compared with  $^{137}\text{Cs}$ , such as  $^{238}\text{Pu}$ , in determining the dose to biota.

A dynamic model in a regulatory context allows the potential impact of accidents on the environment to be more accurately quantified. This would allow regulators to assess the suitability of future nuclear installations in a more comprehensive way as the impact on biota from accident scenarios can be explicitly considered. This allows regulators and other stakeholders such as conservation organisations to have more confidence that nuclear installations such as those being considered as part of the UK's nuclear new build programme over the next 10-15 years will be built on locations that ensure adverse environmental impacts are as low as reasonably practicable. Dynamic modelling allows nuclear companies to demonstrate they have considered potential emergency situations and the possible impact on the environment. This allows nuclear companies to address issues and implement procedures that are optimal for minimising the absorbed dose to biota.

Current regulatory guidelines for the protection of non-human biota are still not as formalised as for human protection. The scientific framework for protecting the environment from ionising radiation should be based on the framework for protecting humans (Pentreath, 2009). The protection of humans is relatively well defined with recognised standard dose limits and an established protection framework in comparison to environmental protection from ionising radiation. The concept and objective of environmental protection often differs widely between countries. The ICRP is trying to establish the scientific basis for environmental protection. The main focus of the ICRP is to prevent environmental effects at the population level. The need to give advice on the level of protection required to protect the environment given the incomplete scientific knowledge of the effect radiation exposure has at the population level is being addressed through the use of Derived Consideration Reference Levels (DCRL) (Pentreath, 2009). These are being derived through the application of current knowledge of radiation effects on biota to a limited number of representative organisms. Together with the Reference Animal and Plants concept this is being used to help inform management decisions under the three ICRP exposure scenarios and provide a framework for environmental protection that can be updated as new data is collected.

The acute dose required for an effect on mammals is 1-10 Gy but a comparison with other biota is difficult because of differences in experiments and time scales. According to Pentreath (2009) dose rates of 0.1 mGy day<sup>-1</sup> are required for a reasonable chance of some effects in higher vertebrates and pine trees, 1-10 mGy day<sup>-1</sup> for lower invertebrates, and 10-100 mGy day<sup>-1</sup> for various invertebrates. Some more detailed effects are given in the dosimetry section above. Current IAEA recommendations give figures for environmental protection of an acute dose of 0.1 Gy for terrestrial biota and a chronic dose of 1 mGy day<sup>-1</sup> for animals and 10 mGy day<sup>-1</sup> for plants (Delistraty, 2008). A time period of <1 day is seen as the exposure period of an acute scenario while chronic exposure covers a significant portion of an organism's lifetime (IAEA, 1992).

In practice many approaches use a tiered system to screen out sites and organisms of negligible concern (Beresford et al., 2010). This is an iterative process with the initial screening levels designed to be simple with minimal input requirements that applies a conservative assessment level of risk to biota or environments to allow them to be removed from the assessment process with a high degree of confidence. The added level of data requirements for a dynamic model and the added uncertainty from requiring more parameters means dynamic modelling is most suited to the later tiers in assessment models that require a more in-depth risk assessment. The regulatory use of models requires a high degree of confidence. Empirical data for model validation is required to gain confidence in the dynamic modelling approach developed here for it to be considered for use in a regulatory context.

# Conclusions

- A new generic dynamic model for the assessment of risk to terrestrial biota from ionising radiation has been developed for  $^{137}\text{Cs}$
- The model has the potential to be extended for other radionuclides provided other pathways highlighted in this study are included such as sea-to-land transfer for the actinides
- A novel approach for calculating the absorbed dose to vegetation from externally deposited radionuclides is presented in this study
- Two proof-of-concept scenarios have been presented to show the assessment capabilities of the dynamic model
- The model has been calibrated to the conditions of a coastal heath dune ecosystem but can be updated and adapted as new data becomes available and for different radionuclides and ecosystem scenarios
- Sensitivity analysis shows model output for activity concentration is most sensitive to the Concentration Ratio parameter
- The  $^{137}\text{Cs}$  deposited onto vegetation accounts for a large amount of the activity concentration when the exposure scenario includes a large atmospheric deposition rate
- An external vegetation compartment doesn't have a large effect on vegetation activity concentration for scenarios with a low atmospheric deposition rate
- To have confidence in the model, an independent data set from a heath dune ecosystem is required for model validation

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## Appendix 1: Atmospheric <sup>137</sup>Cs concentrations from British Nuclear Fuels Limited and Sellafield Limited environmental monitoring reports

The data presented here is the atmospheric concentration of <sup>137</sup>Cs published in the yearly environmental monitoring reports of British Nuclear Fuels Limited and Sellafield Limited. This data is from a location that is south of the Sellafield nuclear facility just outside the perimeter fence.

*Table A1-1: Atmospheric concentration of <sup>137</sup>Cs for each year from 1986-2006 as recorded in Sellafield and British Nuclear Fuels environmental monitoring reports measured from a location close to Sellafield nuclear facility just outside the south site perimeter.*

Year	Atmospheric concentration of <sup>137</sup> Cs from Sellafield south site perimeter (mBq m <sup>-3</sup> )
1986	5.00E+00
1987	7.09E-02
1988	4.35E-02
1989	3.55E-02
1990	8.50E-02
1991	7.62E-02
1992	2.70E-02
1993	1.05E-02
1994	9.00E-03
1995	1.00E-03
1996	1.00E-02
1997	1.00E-02
1998	8.75E-03
1999	3.00E-02
2000	7.00E-03
2001	9.00E-03
2002	8.50E-03
2003	9.00E-03
2004	1.00E-02
2005	1.10E-02
2006	9.00E-03