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Probabilistic biokinetic modelling of radiocaesium uptake in Arctic seal species: verification of modelled data with empirical observations

J. Brown^a, M. Dowdall^b, J.P. Gwynn^b, P. Børretzen^{a,*}, Ø.G. Selnæs^a, K.M. Kovacs^c, C. Lydersen^c

^a Norwegian Radiation Protection Authority, PO Box 55, N-1332 Østerås, Norway ^b Norwegian Radiation Protection Authority, Polar Environmental Centre, N-9296 Tromsø, Norway ^c Norwegian Polar Institute, Polar Environmental Centre, N-9296 Tromsø, Norway

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Abstract

The necessity to provide information about radionuclide concentrations in Arctic marine species has been heightened in recent years due to a number of accidents in Arctic regions involving nuclear vessels and the presence of a large number of potential radioactive contamination sources. The provision for such information is largely dependant on the use of radionuclide uptake and transfer models. The uptake of radionuclides in Arctic seal species in this study has been modelled using a probabilistic biokinetic approach. In this paper, model results are compared with empirical data from relevant samples taken within the Arctic region. Results indicate that the model performs well when estimating concentrations of ¹³⁷Cs in two seal species for both median values and reproduction of the distribution of data values, but not as well for a third seal species. Likely factors affecting the results are the probability density functions used for the input parameters.

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* Corresponding author. Tel.: +47 67 16 26 01; fax: +47 67 14 74 07. *E-mail address:* peer.borretzen@nrpa.no (P. Børretzen).

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1. Introduction

In recent years, substantial focus has been placed upon the development of frameworks and methodologies for assessing the impact of ionising radiation in the environment (e.g. Copplestone et al., 2001; USDoE, 2002; Larsson, 2004). An essential component of any such system is a robust method for deriving radionuclide activity concentrations in the main compartments of an ecosystem. With respect to the biological components of the ecosystem, commonly used methods involve the use of concentration factors (see Harrison, 1986; Gomez et al., 1991), CFs, which can be simply defined as the quotient of the activity concentration of a selected radionuclide in biota (normally $Bg kg^{-1} f.w.$) and the activity concentration in water (normally Bql^{-1} filtered). Such approaches have been widely used in human radiological protection (IAEA, 1985, 2004) and the approach appears to be useful for comparisons and predictions following a suitably long equilibration period (Whicker and Schultz, 1982). However, in situations where activity concentrations are changing rapidly and/or high trophic levels are being considered that may exhibit a slow response to changes in ambient contamination levels, other approaches may be more appropriate. In some cases such as the extrapolation of CF values from acute to chronic contamination situations, from open ocean to coastal sites the approach has been condemned as being highly unsatisfactory (Coughtrey and Thorne, 1983). For this reason, more mechanistically based dynamic models have been applied and, in some cases, tested in various aquatic systems (Thomann, 1981; Kryshev and Ryabov, 2000; Kryshev, 2002).

More recently, a model has been developed for the consideration of High Arctic marine foodchains with some focus on seals (Brown et al., 2004). High trophic level mammals, such as seals are likely to form one important assessment endpoint, inter alia, for any environmental impact assessment owing to their relative radiosensitivity and potential to accumulate certain radionuclides (Hosseini et al., 2005). Ongoing monitoring and research work on seals in the Svalbard region (Gwynn et al., 2005) have provided an excellent opportunity with which to explore and test these models. Furthermore, model functionality pertaining to probabilistic methods and the availability of relatively comprehensive data sets on some of the parameters necessary for the implementation of such simulations has allowed variability and uncertainty to be explored. Probabilistic methods such as Monte Carlo have long been recognised as being useful in efforts to assess and report uncertainty and variability in environmental (in the cited case more specifically human) exposure (Taylor, 1993). Furthermore, the application of probabilistic assessments has been recommended in recently developed systems for assessing the environmental impact of radiation (Brown et al., 2003b) and in the process of establishing the sensitivity of output to parameter variation and the propagation of parameter uncertainty for concomitant models (Avila et al., 2004). Such an approach develops the assessment from a best estimate approach towards a full-fledged risk assessment. The objective of this study was the application of such a probabilistic model to the estimation of ¹³⁷Cs levels in Arctic seal species and a comparison of the model results with available measured data. To the authors' knowledge, this is the first time such a method has been applied in the context of an environmental exposure assessment in the High Arctic.

2. Material and methods

2.1. Studied seal species and model parameterisation

The three species studied are Arctic seal species typically found both on and around Svalbard. The ringed seal (*Phoca hispida*) is the smallest of the seal species, with an adult mean length of about

130 cm and mean adult body mass between 50 and 90 kg (Lydersen and Gjertz, 1987). The most abundant and widely distributed seal species in the northern hemisphere (Reeves, 1998), ringed seals are also the most abundant seal on the Svalbard archipelago, with an estimated population size of more than a hundred thousand (Lydersen, 1998). These seals occur in the archipelago all year round, but are most numerous during winter to early summer when there is ice in the fjords and bays. Ringed seals are opportunistic feeders, preying on a wide variety of pelagic, benthic and ice associated fauna (Gjertz and Lydersen, 1986; Weslawski et al., 1994). The bearded seal (Erignathus barbatus) is the largest of the northern phocid seals, with adult lengths of about 230 cm and weighing on an average 270–275 kg (Andersen et al., 1999), with maximum recordings well over 400 kg. Bearded seals are found in areas of relatively shallow water (<100 m), avoiding areas of continuous fast-ice and are commonly found alongside leads in ice-covered areas or on drifting floes. The circumpolar population display some degree of seasonal movements mainly related to the distribution of sea-ice. On Svalbard, bearded seals are common all year around, throughout the archipelago, with a population probably in the thousands. These seals are predominantly benthic feeders, preving mainly on fish, molluscs and crustaceans (Hielset et al., 1999). The hooded seal (Cystophora cristata) is a pelagic, deep diving seal species, where females can reach 220 cm in length and weigh on an average 200 kg, while adult males can reach 250 cm and 300-400 kg (Kovacs, 2000). Hooded seals can be found off-shore and in areas of broken drift-ice in the North Atlantic and are commonly found off the south-western coast of Spitsbergen in early and later summer and occasionally north of Spitsbergen and Nordaustlandet. The global population is thought to be at least half a million animals. Hooded seal diet consists of a variety of deep-water fish species including Greenland halibut, various redfish species in addition to squid and benthic invertebrates (Kovacs, 2000).

For modelling purposes, standardised dietary compositions based upon Pauly et al. (1998) were used for each seal species. In their approach, diets were apportioned among eight categories of prey types (benthic invertebrates, large zooplankton, small squids, large squids, small pelagics, mesopelagics, miscellaneous fishes and higher invertebrates) and based on published accounts of stomach contents as well as from morphological, behavioural and other information. The modelling results are compared to empirical data derived from the direct measurement of muscle samples of the studied seal species that were collected from the Svalbard region in 2000-2003 (Gwynn et al., 2005). In that study, seal muscle and other tissue samples were obtained by the Norwegian Polar Institute from animals collected for ongoing research programmes (ringed and bearded seals) or through commercial hunting (hooded seals). All tissue samples were stored, frozen at -20 °C prior to analyses. Tissue samples were thereafter cleaned of excess blood, fat and bone fragments and then dried at 105 °C for 24 h or to constant mass. All dried samples were thereafter homogenised using a stainless steel blender, sieved and packed into standardised counting geometries. Samples were counted for the derivation of activity concentrations for gamma-emitting radionuclides using an electrically cooled ptype coaxial high purity germanium detector constructed of low background material (resolution and efficiency of system at 1332 keV was 1.9 keV and 40%, respectively). All measurements were made at the Norwegian Radiation Protection Authority's research unit in Tromsø and subject to the standard QA procedures at that laboratory.

Comparisons between empirical data from Gwynn et al. (2005) and outputs from the model described in this paper will allow us to determine the accuracy of this type of modelling approach in predicting levels of radiocaesium in various seal species.

For the purpose of clarity, the model used in this study is introduced with reference to the ringed seal, modifications of the model for the other two species being presented afterwards.

The following information was utilised with regard to the physical attributes and diet of ringed seals: mean body mass -50-90 kg (Lydersen and Gjertz, 1987); assumed diet -20% benthic invertebrates, 20% large zooplankton, 15% small pelagics, 5%, mesopelagics and 40% miscellaneous fishes (Pauly et al., 1998). These data were used to construct and parameterise a biokinetic model that may be used to predict activity concentrations in the whole-body of ringed seal. The food-chain model used in this study has been modified from the earlier work conducted by Brown et al. (2004). The food-chain structure modelled in this study is shown in Fig. 1.



Fig. 1. Diagrammatic representation of the food-chain model for ringed seals.

2.2. Food-chain model, allometry and probabilistic considerations

The following equations represent the food-chain for the ringed seal:

Trophic level 1: Phytoplankton (equilibrium with water concentration is assumed):

$$C_{\rm p} = CF_{\rm p}C_{\rm w} \tag{1}$$

where C_p is the radionuclide activity concentration in phytoplankton (Bq kg⁻¹ f.w.); CF_p is the concentration factor for phytoplankton (l kg⁻¹); and C_w is the radionuclide activity concentration in seawater (Bq l⁻¹).

Trophic level 2:

Benthic invertebrates

 $C_{\rm bi} = CF_{\rm bi}C_{\rm w} \tag{2}$

where C_{bi} is the radionuclide activity concentration in benthic invertebrates (Bq kg⁻¹ f.w.); CF_{bi} is the concentration factor for benthic invertebrates (l kg⁻¹); and C_w is the radionuclide activity concentration in sea water (Bq l⁻¹).

Zooplankton (uptake via water and food)

$$\frac{\mathrm{d}C_z}{\mathrm{d}t} = \mathrm{AE}_z \cdot \mathrm{IR}_z C_\mathrm{p} + k_{\mathrm{u}z} C_\mathrm{w} - C_z k_{\mathrm{e}z} \tag{3}$$

where AE_z is the assimilation efficiency (dimensionless) for zooplankton; IR_z is the ingestion rate per unit mass of zooplankton (kg f.w. d⁻¹ per kg f.w.); C_p is the activity concentration in phytoplankton (Bq kg⁻¹ f.w.); k_{uz} is the uptake rate of radionuclide to zooplankton directly from water column (d⁻¹); C_w is the activity concentration in water (Bq l1⁻¹); C_z is the activity concentration in zooplankton (Bq kg⁻¹ f.w.); and k_{ez} is the depuration rate from zooplankton (d⁻¹).

Trophic level 3: Fish (uptake via water and food):

$$\frac{\mathrm{d}C_{\mathrm{f}}}{\mathrm{d}t} = \mathrm{AE}_{\mathrm{f}} \cdot \mathrm{IR}_{\mathrm{f}}C_{\mathrm{z}} + k_{\mathrm{uf}}C_{\mathrm{w}} - C_{\mathrm{f}}k_{\mathrm{ef}} \tag{4}$$

where AE_f is the assimilation efficiency (dimensionless) for fish; IR_f is the ingestion rate per unit mass of fish (kg f.w. d⁻¹ per kg f.w.); k_{uf} is the uptake rate of radionuclide to fish directly from water column (d⁻¹); C_f is the activity concentration in fish (Bq kg⁻¹ f.w.); and k_{ef} is the depuration rate from fish (d⁻¹).

Trophic level 4: Ringed seal (uptake via food only):

It was assumed that the uptake of radionuclides directly from the water column to the seals (all species) is negligible. The ringed seal's diet, in simplified terms, consists of 40% fish and 40% zooplankton and 20% benthic invertebrates (see below).

$$\frac{\mathrm{d}C_{\mathrm{rs}}}{\mathrm{d}t} = 0.4(\mathrm{AE}_{\mathrm{rs}} \cdot \mathrm{IR}_{\mathrm{rs}}C_{\mathrm{z}}) + 0.4(\mathrm{AE}_{\mathrm{rs}} \cdot \mathrm{IR}_{\mathrm{rs}}C_{\mathrm{f}}) + 0.2(\mathrm{AE}_{\mathrm{rs}} \cdot \mathrm{IR}_{\mathrm{rs}}C_{\mathrm{bi}}) - C_{\mathrm{rs}}k_{\mathrm{ers}}$$
(5)

where AE_{rs} is the assimilation efficiency (dimensionless) for ringed seal; IR_{rs} is the ingestion rate per unit mass of ringed seal (kg f.w. d⁻¹ per kg f.w.); C_{bi} is the activity concentration in benthic invertebrates (Bq kg⁻¹ f.w.); C_{rs} is the activity concentration in ringed seal (Bq kg⁻¹ f.w.); and k_{ers} is the depuration rate from ringed seal (d⁻¹). All other terms are as defined previously.

Sea water ¹³⁷Cs activity concentrations for the Svalbard region in 2000–2003 ranged from 1.9 to 2.5 mBq l⁻¹ (Gäfvert et al., 2003; NRPA, 2004; NRPA, unpublished data), but for the purposes of modelling a constant ¹³⁷Cs water concentration of 2 mBq l⁻¹ was assumed throughout the course of the simulation. In view of the lack of more explicit information beyond "small" and meso-"pelagics" these organisms have been assumed to be represented by zooplankton with respect to parameters of pertinence to the model for all three species studied. It has furthermore been assumed that the category fish, forming part of the seal's diet, covers a spectrum from small (0.005–50 g f.w.), trophic level 3, to large (0.5–5 kg f.w.), trophic level 4 fish.

With respect to the depuration rates of ¹³⁷Cs from ringed seal, two approaches have been adopted. One is based on the allometric relationship defined by Whicker and Schultz (1982) and expressed in Eq. (6) below. A mass of 50 kg, i.e. the lowest mass in the range given for adult ringed seals, yielding the highest depuration rate, was use in this formula to derive the highest values in the range for the parameter value $k_{\rm ers}$.

$$\lambda_i = \frac{\ln 2}{18.36M^{0.24}} \tag{6}$$

where λ_i is the depuration rate (d⁻¹); *M* is the mass of organism in kg.

The other method is based on the retention functions defined by ICRP (1979) for adult man (Eq. (7)). Although radiocaesium is known to deposit mainly in mammalian muscle and the muscle/body, mass ratio may be quite different in seals and man thereby affecting the excretion kinetics, the use of human retention functions was considered to provide a reasonable estimation for radiocaesium depuration rates from the body of seals (notably with lean body masses similar to those for adult humans). Used in combination with other information these human data help us define bounds within which the loss of radiocaesium from seals is likely to fall,

$$R(t) = 0.1e^{-0.693t/2} + 0.9e^{-0.693t/110}$$
(7)

where R(t) is the radionuclide retention.

Taking the long component of the retention function for man a k_{ers} of $6.3 \times 10^{-3} d^{-1}$ is derived. This value was used to set the lower value of the range used for this parameter.

Concentration factors have been employed for both phytoplankton and benthic invertebrates. The implicit assumption behind this is that the water compartment and biotic compartment equilibrate instantaneously. This assumption appears valid for some radionuclides and phytoplankton as discussed elsewhere. The assumption may be less valid for benthic invertebrates that often derive their food from organic detritus, e.g. consider the case of bivalve molluscs, which may or may not be in equilibrium with the ambient water. With respect to the CFs employed for benthic invertebrates, it has been assumed that values for benthic molluscs will provide a reasonable representation of transfer to this broad class of organism. Studies by Rissanen et al. (1997) considering a large spectrum of various benthic invertebrate species from northern sea areas would appear to substantiate this view.

It has also been assumed that the mass of the animals considered in this simulation is at steady state. First order growth rate constants, as applied in some models to account for biomass increases (see Landrum et al., 1992) have therefore not been applied. An overview of the parameters used is provided in Table 1. Similar simulations were also carried out for hooded and bearded seal based partly on information provided with regard to the mammals' mass and diet. Bearded and hooded seals are much larger mammals than ringed seals with adult body masses around 270–275 kg (Andersen et al., 1999) and 200–400 kg (Kovacs, 2000), respectively. According to scaling laws, i.e. allometry, this is likely to influence several parameters including ingestion rates and ¹³⁷Cs depuration rates. For this reason some modifications were made to the parameters used for ringed seal. Applying the appropriate allometric relationship pertaining to ingestion rates for seals IR = $0.079M^{0.71}$, where *M* is the mass in kg, a mass-normalised IR of 0.0155 kg f.w. d⁻¹ per kg f.w. was obtained. Since no variation in adult sizes is reported an arbitrary ±20% has been assigned to the adult mass to derive a range in expected IR values (see Table 2).

With respect to 137 Cs depuration rates for both hooded and bearded seals, parameter values have been derived using appropriate allometric relationships. In addition, the long period component of retention functions for man has been modified in correspondence with the changes observed for the allometrically derived depuration rates. More specifically – application of allometric relationship from Eq. (6) gives a 137 Cs depuration rate of 9.8×10^{-3} (d⁻¹) for a 275 kg mammal. This value compares to a representative value of 0.0136 derived for ringed seal (of mass 70 kg) based on allometry alone, i.e. the value for hooded and bearded seal is 0.72 times the value for ringed seal. Applying this same factor to the retention function value of 6.3×10^{-3} yields a value of 4.5×10^{-3} (d⁻¹).

The dietary composition assumed for bearded seals was 65% benthic invertebrates, 15% large zooplankton, 5% small pelagics, 15% miscellaneous fishes (Pauly et al., 1998). This has been accounted for by weighting the food-chain diagram (see Fig. 1) and relevant equations accordingly. Eq. (5) for ringed seals thus becomes Eq. (8) for bearded seals employing the same assumptions as for ringed seals:

$$\frac{\mathrm{d}C_{\mathrm{bs}}}{\mathrm{d}t} = 0.2(\mathrm{AE}_{\mathrm{bs}}\cdot\mathrm{IR}_{\mathrm{bs}}C_{\mathrm{z}}) + 0.15(\mathrm{AE}_{\mathrm{bs}}\cdot\mathrm{IR}_{\mathrm{bs}}C_{\mathrm{f}}) + 0.65(\mathrm{AE}_{\mathrm{bs}}\cdot\mathrm{IR}_{\mathrm{bs}}C_{\mathrm{bi}}) - C_{\mathrm{bs}}k_{\mathrm{cbs}} \tag{8}$$

where AE_{bs} is the assimilation efficiency (dimensionless) for bearded seal; IR_{bs} is the ingestion rate per unit mass of bearded seal (kg f.w. d⁻¹ per kg f.w.); C_{bi} is the activity concentration in benthic invertebrates (Bq kg⁻¹ f.w.); C_{bs} is the activity concentration in bearded seal (Bq kg⁻¹ f.w.); and k_{ebs} is the depuration rate from bearded seal (d⁻¹).

The dietary composition assumed for hooded seals was; 20% small squids, 20% large squids, 20% small pelagics, 40% miscellaneous fishes (Pauly et al., 1998). Cs-137 CF values are available for cephalopods in IAEA (2004). The relevant value of 9 with a concomitant range of ± 1 order of magnitude has been drawn from this reference. The pathway from water to benthic invertebrates as shown in Fig. 1 is now replaced by a pathway from water to squid. Eq. (5) for ringed seals is modified to become Eq. (9):

$$\frac{\mathrm{d}C_{\mathrm{hs}}}{\mathrm{d}t} = 0.2(\mathrm{A}\mathrm{E}_{\mathrm{hs}}\cdot\mathrm{I}\mathrm{R}_{\mathrm{hs}}C_{\mathrm{z}}) + 0.4(\mathrm{A}\mathrm{E}_{\mathrm{hs}}\cdot\mathrm{I}\mathrm{R}_{\mathrm{hs}}C_{\mathrm{f}}) + 0.4(\mathrm{A}\mathrm{E}_{\mathrm{hs}}\cdot\mathrm{I}\mathrm{R}_{\mathrm{hs}}C_{\mathrm{c}}) - C_{\mathrm{hs}}k_{\mathrm{ehs}}$$
(9)

where AE_{hs} is the assimilation efficiency (dimensionless) for hooded seal; IR_{hs} is the ingestion rate per unit mass of hooded seal (kg f.w. d⁻¹ per kg f.w.); C_c is the activity concentration in cephalopods (Bq kg⁻¹ f.w.); C_{hs} is the activity concentration in hooded seal (Bq kg⁻¹ f.w.); k_{ehs} is the depuration rate from hooded seal (d⁻¹).

The equations representing the systems as described above were solved numerically using the software ECOLEGO run within a Matlab[©] environment (Avila et al., 2003). The model was run probabilistically using a Monte Carlo simulation of 1000 iterations.

Parameter	Description (units)	Distribution	Value (min, max)	Reference(s)	
AEz	Assimilation efficiency of ¹³⁷ Cs in zooplankton (dimensionless)	Uniform	0.75 (0.5, 1)	Thomann (1981), USDoE (2002)	
AE_{f}	Assimilation efficiency of ¹³⁷ Cs in fish (dimensionless)	Uniform	0.75 (0.5, 1)	Thomann (1981), USDoE (2002)	
AE _{rs}	Assimilation efficiency of ¹³⁷ Cs in ringed seal	Uniform	0.75 (0.5, 1)	Thomann (1981), USDoE (2002)	
CF _p	Concentration factor for phytoplankton (1 kg^{-1})	Triangular	20 ^a (1, 100)	IAEA (2004)	
CF _{bi}	Concentration factor for benthic invertebrates taken to equate to that for mollusc (1 kg^{-1})	Triangular	50 ^a (5, 500)	Brown et al. (2003a)	
IRz	Ingestion rate for zooplankton $(\text{kg d}^{-1} \text{ per kg f.w.})$	Uniform	0.105 (0.084, 0.126) ^b	Thomann (1981)	
IR_{f}	Ingestion rate for fish $(\text{kg d}^{-1} \text{ per kg f.w.})$	Uniform	0.013 (0.009, 0.017) ^c	Thomann (1981)	
IR _{rs}	Ingestion rate for ringed seal (kg d^{-1} per kg f.w.)	Uniform	0.023 ^d (0.0214, 0.0254)	Innes et al. (1987)	
k _{ez}	The depuration rate of 137 Cs from zooplankton (d ⁻¹)	Uniform	0.03 (0.024, 0.036) ^b	Thomann (1981)	
k _{ef}	The depuration rate of 137 Cs from fish (d ⁻¹)	Uniform	0.006 (0.0018, 0.01) ^c	Thomann (1981)	
<i>k</i> ers	The depuration rate of 137 Cs from ringed seal (d ⁻¹)	Uniform	0.01 (0.0063, 0.0148) ^e	ICRP (1979), Whicker and Schultz (1982)	
k _{uz}	Rate of 137 Cs uptake by zooplankton directly from the water column (d ⁻¹)	Uniform	0.49 (0.39, 0.59) ^b	Thomann (1981)	
k _{uf}	Rate of 137 Cs uptake by fish directly from the water column (d ⁻¹)	Uniform	0.04 (0.01, 0.07)	Thomann (1981)	

Parameters used in the probabilistic assessment for transfer of ¹³⁷Cs to ringed seals

Table 1

^a For triangular distributions the value given is the mode.

^b Range estimate by deriving $\pm 20\%$ of recommended parameter value.

^c Range expressed by parameter values for small and large fish as reported by Thomann (1981).

^d Allometrically derived, based on range in masses of 50–90 kg.

^e Range derived from long period retention function for man (ICRP, 1979) and allometric value for a 50 kg seal from Whicker and Schultz (1982).

3. Results and discussion

Reported ¹³⁷Cs activity concentrations in muscle and summary statistics for the simulation results for the three seal species are detailed in Table 3. Outputs from the model runs for ringed seals are displayed in Figs. 2 and 3, the output from the model being used to predict a median ¹³⁷Cs activity concentration in ringed seal of approximately 0.52 Bq kg⁻¹ with a concomitant range (this being arbitrarily defined in this study as the range between the 25th and 75th percentile) of 0.36–0.8 Bq kg⁻¹ f.w. This compares to a range (between 25th and 75th percentile) of 0.44–0.55 Bq kg⁻¹ f.w. from empirical observation. In the model, the time taken for a ringed seal to reach near equilibrium with ambient seawater ¹³⁷Cs concentrations is approximately 500

Parameter	Description (units)	Distribution	Value (min, max)	Reference(s)
IR _{bs} IR _{hs}	Ingestion rate for bearded or hooded seal $(kg d^{-1} per kg f.w.)$	Uniform	0.0155 (0.0124; 0.0186)	Innes et al. (1987)
$k_{ m ebs}$ $k_{ m ehs}$	The depuration rate of ¹³⁷ Cs for bearded	Uniform	7.15×10^{-3} $(4.5 \times 10^{-3}; 9.8 \times 10^{-3})$	ICRP (1979), Whicker and Schultz (1982)
CF _{ceph}	or hooded seal (d^{-1}) Concentration factor for cephalopods $(l kg^{-1})$	Triangular	9 (0.9, 90)	IAEA (2004)

Modified parameters from Table 1 for use in the food-chain simulations for bearded and hooded seals

days. It should be noted that the empirical data for seal species is unlikely to represent equilibrium conditions due to seasonal variations in prey availability and individual feeding rates as well as the additional impact of migration patterns across areas of variable seawater ¹³⁷Cs concentrations.

Predicted activity concentrations for bearded seals (Figs. 4 and 5) are similar to those for ringed seal, although the prognosis for the maximum value in the range lies slightly below that for ringed seal. The model output for ¹³⁷Cs activity concentrations in bearded seal, at equilibrium, yields a median value of 0.55 Bq kg⁻¹ and a range (defined by the lower 25th and upper 75th percentile) of 0.35–0.80 Bq kg⁻¹. This compares to empirical data expressing a range (defined by the 25th and 75th percentile) of 0.15–0.29 Bq kg⁻¹. The ¹³⁷Cs activity concentrations in hooded seal (Figs. 6 and 7) predicted from model simulation, median 0.41 Bq kg⁻¹ and range, defined by 25th and 75th percentiles, 0.28–0.65 Bq kg⁻¹ compare to the empirical data expressing a commensurable range of 0.28–0.36 Bq kg⁻¹. The model successfully predicts the slightly lower activity concentrations as observed for hooded seals, this appearing to reflect the significant proportion of cephalopods in the diet of this seal species, an organism that is known to bio-concentrate ¹³⁷Cs to a lower level than other marine organism groups including molluscs and fish. Indeed, Yoshitome et al. (2003) reported an inverse relationship between ¹³⁷Cs concentration factors and the percentage of cephalopods in the diet of various marine mammals.

Median values for modelled data and empirical observations were compared statistically (Mann–Whitney) in order to ascertain whether significant differences could be said to exist between the two sets. No significant difference could be observed for the ringed (P = 0.285) or

	Ringed seal		Bearded seal		Hooded seal	
	Model	Empirical	Model	Empirical	Model	Empirical
n	1000	15	1000	7	1000	10
Mean	0.646	0.488	0.634	0.222	0.522	0.317
Std. Dev.	0.405	0.072	0.362	0.124	0.398	0.073
Min	0.102	0.400	0.078	0.061	0.077	< 0.2
25%	0.361	0.440	0.353	0.152	0.280	0.281
Median	0.521	0.477	0.550	0.191	0.408	0.324
75%	0.803	0.551	0.797	0.293	0.647	0.355
Max	3.122	0.610	2.321	0.416	3.301	0.418
Skewness	1.590	0.497	1.296	0.555	2.722	-0.244
Kurtosis	3.319	-1.041	2.007	-0.543	11.367	-0.022

Comparison of summary statistics for model outputs and empirical data (Gwynn et al., 2005)

All values are 137 Cs in Bq kg $^{-1}$ (f.w.).

Table 2

Table 3



Fig. 2. Cs-137 activity concentration in ringed seal versus time. The values plotted are for the median (0.52 Bq kg⁻¹), 25th (0.36 Bq kg⁻¹) and 75th percentile (0.80 Bq kg⁻¹).

hooded (P = 0.155) seal species. The situation was not the same for the bearded seal where the modelled and actual data displayed significant differences (P = 0.001). The distribution of the modelled results for the bearded seal species differs slightly to those of the hooded and ringed species in that the skew towards higher values is caused by a larger number of data points than for the latter two (cf. Fig. 5 with Figs. 3 and 7). Assuming the probabilistic modelled data are considered to be representative for the models themselves, the validity of the models was then investigated by comparing the modelled data with the empirical data, using the non-parametric Kolmogorov–Smirnov test for goodness of fit (Massey, 1951), the results of this analysis being presented in Figs. 8–10. This test compares the differences in cumulative probabilities between



Fig. 3. Histogram showing output in terms of 137 Cs activity concentrations in ringed seal at time = 5000 days, i.e. at equilibrium.



Fig. 4. Cs-137 activity concentration in bearded seal versus time. The values plotted are for the median (0.55 Bq kg⁻¹ at equilibrium), 25th (0.35 Bq kg⁻¹ at equilibrium) and 75th percentile (0.80 Bq kg⁻¹ at equilibrium).

the two investigated sets of data, and this difference is considered statistically significant if it exceeds a critical value $d_{\alpha,\text{model}}$. At the $\alpha = 0.01$ level of significance, this value is defined as $d_{0.01,\text{ringed}} = 0.418$, $d_{0.01,\text{bearded}} = 0.618$ and $d_{0.01,\text{hooded}} = 0.490$, for the ringed, bearded and hooded seals data, respectively. At this level of significance, there were found no significant differences between the modelled and empirical data for ringed and hooded seals. However, for bearded seals, a significant difference was again observed.

The differences observed between modelled and empirical data for bearded seals may partly be explained by the use of estimated dietary compositions, which in reality will vary from



Fig. 5. Histogram showing output in terms of 137 Cs activity concentrations in bearded seal at time = 5000 days, i.e. at equilibrium.



Fig. 6. Cs-137 activity concentration in hooded seal versus time. The values plotted are for the median (0.4 Bq kg⁻¹ at equilibrium), 25th (0.27 Bq kg⁻¹ at equilibrium) and 75th percentile (0.62 Bq kg⁻¹ at equilibrium).

location to location. The use of more accurate and site specific dietary information may improve the model predictions for this species, as well as for the other seal species in this study.

With respect to the higher values that skew the distribution of the modelled results, the distributions attributed to each of the parameters used in the assessments warrant some attention. According to Taylor (1993), uncertainty may be considered as a measure of (i) the incompleteness of one's knowledge or (ii) information about a quantity whose true value might be established was a perfect measuring device available. Variability, on the other hand, relates to temporal, spatial or inter-individual heterogeneity in the input/parameter value. From these definitions, all parameter distributions provided in this work can be essentially considered as



Fig. 7. Histogram showing output in terms of 137 Cs activity concentrations in hooded seal at time = 5000 days, i.e. at equilibrium.



Fig. 8. Cumulative probability plots for modelled and measured ringed seal data (Gwynn et al., 2005). Limits plotted at the 0.01 significance level.

combined uncertainty and variability distributions. In an ideal situation, large empirical data sets would be available for each of the parameters of interest to allow detailed histograms to be plotted and the most appropriate probability density function to be assigned, whether that is log-normal, normal, Weibull, etc. In most cases, lack of empirical data forces a more judgemental approach to the generation of a suitable distribution. In the assessment described in the present paper, available information was often minimal with little data beyond a range reported. In this case a uniform distribution was assumed to be a reasonable choice wherein all values within the defined range have an equal likelihood of occurrence. In some instances, such as data pertaining to concentration factors for low trophic level organisms, the information



Fig. 9. Cumulative probability plots for modelled and measured bearded seal data (Gwynn et al., 2005). Limits plotted at the 0.01 significance level.



Fig. 10. Cumulative probability plots for modelled and measured hooded seal data (Gwynn et al., 2005). Limits plotted at the 0.01 significance level.

coverage was considered sufficient to justify the employment of a triangular distribution whereby a mid-value is deemed most likely - in these few cases the peak was identical to the best estimate CF values identified by IAEA (2004) for these organism groups. The cause of the skewed distribution observed in the model results may therefore be a consequence of the distributions used for the inputs.

Fowler and Fisher (2004) have noted that there is conflicting evidence regarding the observation of whether biomagnification occurs at the top of marine food-chains or not. Whereas Watson et al. (1999) reported that seals from the Irish Sea contained ¹³⁷Cs concentrations some 3-4 times greater than those of local fish, their main food source, seals in the Arctic showed little difference between their ¹³⁷Cs concentrations and those of their prey (Carroll et al., 2002). The present study may elucidate this situation to some extent in that the model indicates that seal body concentrations appear to be very sensitive to the contamination levels and relative proportions of prey types constituting the seal's diet. In the case of ringed seal, for example, biomagnification appears to be quite evident with this species of seal exhibiting body activity concentrations that are 1.5-2.5 times greater than the main prey species, i.e. fish and zooplankton (Fig. 11a). In contrast, hooded seal actually exhibit activity concentrations that fall slightly below fish activity concentrations that form an important part of their diet. This reflects the fact that cephalopods, with relatively low contamination levels, are an equally important part of the seal diet and result in lowered body burdens of ¹³⁷Cs in hooded seal (Fig. 11b). The question of biomagnification cannot therefore be addressed without considering in detail the diet of the studied animal taking due consideration of variability inter- and intra-species.

4. Conclusions

Cursory inspection of data suggests that the models predict empirical values fairly robustly for all three seal species – model predictions fall well within 1 order of magnitude of the actual measurements. Non-parametric statistical tests to determine whether there were significant differences between empirical and model data sets and to establish how closely empirical and



Fig. 11. Activity concentrations for different trophic levels based on model simulation for (a) ringed seal and prey species and (b) hooded seal and prey species.

model probability distributions correspond confirmed that there were no significant differences in the cases of ringed and hooded seals. For bearded seal, however, differences were significant. The reason why the model predictions for bearded seal are relatively poor is unclear but may relate to that large proportion of benthic invertebrates in the diet of this seal species and the concomitant uncertainties associated with contamination levels in this dietary component. For the sake of simplicity the default concentration factor for benthic invertebrates equate to that for mollusc, which may not be especially representative of the actual benthic invertebrates forming the seals diet.

Further refinement of the models could be achieved by refining parameters in such a way that the probability distribution associated with each might be represented in a more realistic fashion than the ubiquitously used "uniform" distribution. In some cases this would require extensive experimental study, as exemplified by the study of assimilation efficiencies and depuration rates in organisms. Other improvements might be introduced by including a growth rate within the equations describing the system although in the case of seals, this might only improve predictions for actively growing juvenile animals. Parameterisation might, in any case, be difficult here. Finally, in view of the dynamic nature of the model, the assumption that ambient seawater and hence prey concentrations have remained at a constant level over the simulation period of several thousand days may be slightly erroneous. In reality, seals are exposed to a range of ambient seawater concentrations due to a combination of the seal's migratory patterns and the effects of oceanic mixing.

The biokinetic approach does provide more precise predictions of the activity concentrations in seals compared to a simpler approach based on the application of CFs and furthermore provides differentiation between various seal species reflecting their particular dietary intake. The fully dynamic modelling approach leads to refinement in our mechanistic understanding of this ecological system and we believe that rapidly fluctuating environmental concentrations would lead to a far superior prognosis than that derived from the application of a concentration factor. The approach has undoubted utility within the broad context of environmental impact assessments and with further testing may be appropriately used in order to make prediction following intermittent or accidental release of radionuclides in Arctic ecosystems.

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