

## Environmental impact assessments for the marine environment – transfer and uptake of radionuclides



*Reference:*

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*Key words:*

Marine radioactivity, compartment models, trophic transfer, concentration factors, biokinetic modelling.

*Abstract:*

The first stage of an environmental impact assessment for radionuclides in marine environments is considered within this report. Types of physical process models are described before data pertaining to biota concentration factors are reviewed. In the final section of the report, biokinetic models, which can be used to simulate the biological transfer of radionuclides in northern marine systems, are presented.

*Referanse:*

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*Emneord:*

Marin radioaktivitet, boksmodeller, trofisk overføring, konsentrasjonsfaktorer, biokinetisk modellering

*Resymé:*

Denne rapporten omhandler første trinnet i en miljøkonsekvensanalyse for radionuklider i det marine miljø. Forskjellige havtransportmodeller er beskrevet fulgt av en gjennomgåelse av data for biokonsentrasjonsfaktorer. I den siste delen av rapporten blir biokinetiske modeller, som kan brukes for å simulere biologisk overføring av radionuklider i nordlige marine systemer, presentert.

Head of project: Justin Brown

*Approved:*



Per Strand, Director, Department for Environmental Protection and Emergency Response

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*Orders to:*

Norwegian Radiation Protection Authority, P.O. Box 55, N-1332 Østerås, Norway.

Telephone +47 67 16 25 00, fax + 47 67 14 74 07.

[www.nrpa.no](http://www.nrpa.no)

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## I. Summary

In order to assess whether the environment is being protected from ionising radiation, a methodology is required for the purpose of predicting activity concentrations in selected “reference” organisms and their habitat. When considering the specific example of the marine environment, the first stage in this type of assessment requires a simulation of the physical dispersion of contaminants, whether by advection and dispersion in the water column or interaction with and subsequent transport by sediment. This report provides an overview of some of the models available for this type of simulation, primarily from European Research groups, and more details on the application of NRPA’s in house model simulation tool, the “NRPA box-model”, in simulating the behaviour of radionuclides in Arctic marine environments. In this special case, interaction with and transport by ice requires some attention.

Following the physical transport of contaminants, transfer to biota in marine systems has traditionally been modelled, in the process of calculating doses to humans, using “Concentration Factors (CFs)” that relate the body concentration, or organ concentration, of a specified organism and radionuclide to the ambient sea water concentration. Such values are normally derived from empirical observations. Notwithstanding the limitations associated with the CF approach, as discussed within this report, new data based on recently-published information have been collated in order to augment standard databases from the perspective of environmental, as opposed to human-relevant, pathways. In addition, an alternative approach to predicting activity concentrations in organisms is demonstrated through the application of simple biokinetic models. These models can be used to predict dynamic concentrations (and CFs) in high trophic level organisms, including for example pinnipeds, empirical radionuclide transfer data for which are often sparse or non-existent. The future application of these models, following more detailed parametrisation and validation, will provide an invaluable tool for the authority in its quest to perform robust, defensible environmental impact assessments.

## II. Sammendrag

For å kunne bestemme om miljøet er beskyttet fra ioniserende stråling, er det nødvendig å ha en metode for å kunne forutsi aktivitetskonsentrasjonen i utvalgte referanseorganismer og deres habitat. Ved å se på det marine miljøet som et spesifikt eksempel, trengs det i en slik vurdering, for det første, en simulering av forurensingsstoffenes fordeling mellom forskjellige faser i havet, dvs i hvilken grad stoffene forflyttes via adveksjons- og dispersjonsprosesser og hvordan de vekselvirker og transporteres med sedimenter. Denne rapporten gir en oversikt over noen av de modeller som er tilgjengelige for denne typen simuleringer (primært fra europeiske forskningsgrupper), og mer detaljert informasjon er gitt om bruken av Strålevernets egne modellsimuleringsverktøy, ”NRPA’s bokmodell”. Denne brukes for å simulere oppførselen til radionuklider i Arktiske marine miljøer. Her er også interaksjonen og transport med is tatt med til en viss grad.

Neste skritt har tradisjonelt sett vært å modellere overførselen til biota og deretter til mennesker (for å beregne doser), ved å bruke konsentrasjonsfaktorer (CF) som relaterer helkropps- eller organkonsentrasjonen i en spesifikk organisme til den omkringliggende vannkonsentrasjonen. Slike tall er normalt sett utledet fra empiriske observasjoner. Til tross for de begrensninger med CF tilnærmingen som er diskutert i denne rapporten, har nye data basert på nylig publisert informasjon blitt samlet inn for å utvide standard databaser med CF for næringskjeder relevante i et miljøperspektiv i motsetning til et menneskesentrert perspektiv. I tillegg er en alternativ tilnærming til å forutsi aktivitetskonsentrasjoner i organismer demonstrert ved å bruke enkle biokinetiske modeller. Disse modellene kan brukes for å forutsi dynamisk varierende konsentrasjoner (og CF) i organismer høyt opp i næringskjeden (for

eksempel seler) som det er lite eller ingen empiriske data tilgjengelig for. Videre utvikling av disse modellene, inkludert en mer detaljert parametrisering og validering, vil gi et uvurderlig verktøy for myndigheter med interesse for å kunne utføre robuste og vitenskapelig forsvarlige miljøkonsekvensanalyser

# 1. Introduction

## 1.1 Background and scope of report

Within the field of radiological protection, it has historically been assumed (ICRP, 1977; ICRP, 1991) that by protecting man from the effects of ionising radiation, the environment is automatically protected. Although this tenet may have some basis for application based on the fact that humans are known to be relatively radiosensitive and that limits to protect man are set at very low levels, evidence to consolidate this belief have never been expressed in concrete terms (Pentreath 1998).

The inclusion of references to the protection of the environment in numerous international conventions, principles and statements of intent (e.g. AEPS, 1991, IAEA, 1995; UNCED, 1992; OSPAR, 1998) has augmented pressure for the introduction of an environmental impact assessment methodology, considerably. Publications exist in the open literature which deal with themes related to the assessment of radioactivity and radiation effects in selected environments (IAEA, 1979, IAEA, 1988; NCRP, 1991, UNSCEAR, 1996). This background information has allowed some countries, already, to take steps in response to environmental protection legislation by providing guidance on environmental impact assessments for ionising radiation (Copplesstone et al., 2001; USDoE, 2002). However, there is currently considerable divergence in structure and content of these methodologies, for example with respect to transfer data and models incorporated, dosimetric models employed and endpoints of concern. For the sake of clarity, it has become increasingly apparent that a structured, internationally recognised framework for assessing the impacts of radioactivity explicitly for the environment, is required. This is a standpoint now advocated by a number of international organisations including the International Commission on Radiological Protection and the International Union of Radioecology.

A number of recent publications (Pentreath 1998; Pentreath, 1999; Strand et al., 2000; Strand & Larsson, 2001) have called for the development a system for protecting the environment from ionising radiation. Such a system would allow the considerable volume of available data to be organised in a systematic manner. Basic, although essential, components of this system would include a reference set of organisms that could act as representative of the larger ecosystem, a set of quantities and units allowing consistent comparison of the effects from different radiation types, a set of dose models to allow calculation of absorbed dose and tabulated dose-effects relationships to allow interpretation of the doses received. Within this system a transparent, defensible impact assessment could be performed.

The term “reference organism” has been defined as : “a series of entities that provides a basis for the estimation of the radiation dose rate to a range of organisms that are typical, or representative, of a contaminated environment. These estimates, in turn, would provide a basis for assessing the likelihood and degree of radiation effects.” (Larsson et al., 2002). Numerous criteria that might be used in the selection of reference organism types have been previously suggested (Pentreath & Woodhead, 2001). A list of the criteria adopted might include :(i) organisms that are typical or ubiquitous (ii) importance for the functioning of the ecosystem (iii) potential for high internal and external exposure from radiation, (iv) the availability of radiobiological information (v) the radiosensitivity of the organism (vi) amenability to future research. These and related criteria have been applied in a fairly simple way in order to identify suitable reference organism types for the marine, as well as the terrestrial, environment in several reports and papers compiled within the FASSET and EPIC projects (see Beresford et al., 2001; Strand et al., 2001; Wright et al., 2002; Sazykina et al., 2002)

Clearly, the scope of the “system” is large and cannot be covered in detail through one report. The initial development of the system as a whole is being undertaken through 2 EC projects Framework for the



Assessment of Environmental Impact - "FASSET" and Environmental Protection from Ionising Contaminants in the Arctic "EPIC" (Larsson et al., 2002). This report will build upon the developments outlined above and concern the initial component of the system, namely a methodology for predicting activity concentrations in biota and their habitat for the specific case of the marine environment. Such data may be subsequently used to calculate internal and external doses to biota through the application of suitable dosimetric models. The environmental dose data may then be interpreted through reference to appropriate dose-effects databases. In many cases, e.g. routine releases, chronic exposure data may be of greatest relevance to the assessment. Furthermore, in view of their significance to population integrity as well as the individual organism, reproductive endpoints (i.e. fertility and fecundity) may form a focal point for data collation and further study. It should be emphasised, however, that dosimetric and dose-effects considerations will not form part of this current report.

## 1.2 Report Structure

The report is split into 5 chapters. Following this introductory section, a consideration of the types of model available for the prediction of radionuclide dispersal and fate in marine environments will be given. Within Chapter 3 standard methods for modelling uptake in aquatic foodchains will be presented along with data collated specifically for the purpose of marine environmental impact assessments. Finally, in Chapter 4, a dynamic modelling approach will be outlined before conclusions are drawn in the final Chapter.

## 1.3 Behaviour and fate of radionuclides in marine systems

Following the release/input of a radionuclide suite to the surface waters of a marine system several immediate processes are likely to occur. A fraction of the radionuclide inventories will be advected by prevailing currents away from the source and diluted and dispersed by diffusion processes. The remaining fraction of the radionuclide inventories will undergo interaction with suspended particulate material and uptake by biota. The degree of particulate-phase interaction will depend on numerous factors including the physico-chemical form of the radionuclide, the availability of adsorption surfaces and the lithology and chemical attributes of the suspended material.

Biological uptake can occur through the process of adsorption onto cell surfaces, e.g. actinide adsorption to phytoplankton cells. Those radionuclides that are analogues/radioisotopes of metals important in enzyme systems, for example, will be actively taken up. Passive uptake may occur following the adsorption of radionuclides/heavy metals on organic particles following interaction with surface groups, e.g. carboxylic, phenolic (Millero, 1996). Those radionuclides that are assimilated to a significant degree by biota in the epipelagic zone will enter the pelagic food-chain. Biomagnification may occur in some instances whereby increasing activity concentrations are associated with successively higher trophic levels. In contrast those radionuclides that have no natural, biologically important analogues may be actively selected against and exhibit reduced activity concentrations at higher trophic levels.

Radionuclides can be removed from the water column and transferred to bottom sediments by several processes including

- (i) Direct uptake of the radionuclide at the sediment-water interface.
- (ii) Sedimentation with organic matter either after assimilation or adsorption onto cell surfaces
- (iii) Adsorption onto inorganic compounds (e.g. clays, carbonates) or scavenging from solution by iron-manganese oxy-hydroxides.
- (iv) Sedimentation with humic matter

The physical and chemical properties of the seawater and sediments along with the physico-chemical form of the radionuclide will regulate the magnitude of each possible mechanism. Grain-size often

strongly influences the activity concentration of radionuclides in marine sediments (Hetherington & Jefferies, 1974; Bonnett et al., 1988; Assinder et al., 1993; Clifton et al., 1997).

In marine environments, conservative radionuclides introduced at the sea surface will migrate downwards by the process of diffusion and advection. (Bowen et al., 1980). The vertical flux of water will influence how quickly the radionuclides will come into contact with bottom sediments. In the deep ocean, advection in the vertical plane may be limited. An example can be found in the Arctic Sea where stratification prevents winter convection and deepening of the polar mixed layer reducing the flow of contaminants to deeper layers (Gregor et al., 1998). In the European shelf seas where water depths are < 500m and the epipelagic layer is prone to the turbulent action of winds and tides, the whole water column often becomes well mixed. Radionuclides that were originally introduced at the surface will be mixed throughout the water column after a relatively short period (months → years). Evidence for this can be derived from the fact that “conservative” radionuclides introduced as global fallout or from W-European reprocessing plants can be observed at enhanced levels in deeper shelf waters (e.g. see Kershaw & Baxter, 1995). The settling velocity of the particles will also influence the removal rate of particle-reactive radionuclides from the water column. Large particles will, according to Stoke’s law, fall rapidly. An important consideration relating to removal mechanisms and rates is the nature of the inorganic and organic components of the suspended matter. It is apparent that the dominant mass flux of matter to the seafloor is by the rapid transport of large particles. These are faecal pellets from grazing zooplankton containing organic matter, skeletal material and minor amounts of clay minerals. Sinking rates of approximately 100 m day<sup>-1</sup> are required to account for the material caught in traps set at abyssal depths (Sholkovitz, 1983).

Once incorporated into sediments, radionuclides will be exposed to early diagenetic reactions. This will either result in the virtually irreversible binding of the radionuclide to robust phases of the sediment matrix or to redissolution and return of radionuclide to the water column. Geochemical phase association studies for example via sequential extractions of sediments provide information with regards to these processes.

The process of physical disturbance and bioturbation leads to the mixing of radionuclides in the surface layer of the sediment over short time periods. In the north East Irish Sea for example, mixing of surface sediment (< 13 cm) occurs on a time-scale of ca. 1 year (Mackenzie et al., 1998). Biological activity in this area is both extensive and heterogeneous and is probably responsible for the great variety of vertical profiles of Sellafield radionuclides which have been observed in cores taken from the Irish Sea (Kershaw et al., 1992). The sedimentation of particulate material will also lead to the burial of contamination. The net sedimentation rate is approximately 0.1 mm y<sup>-1</sup> for the NE Irish Sea (Kershaw et al., 1992) but is in the order of mm per year (Brown *et al.*, 1999a) for coastal/estuarine environments in the same area. Sedimentation rates in the open ocean may be a thousand times lower than for coastal deposits (Gregor *et al.*, 1998). Contaminated sediments are also prone to resuspension and may be subsequently transported by prevailing currents. A third process leading to the redistribution in sediment is the dissolution and vertical migration of radionuclides *via* pore waters.

In addition to physical-chemical interaction, direct uptake through assimilation by biota on bottom sediments may also occur. Suspension feeders extract particles from the water column and in so doing also ingest contaminants. Radionuclides will also be introduced to the benthic foodchain via direct uptake, or adsorption, to benthic primary producers such as macroalgae and benthic diatoms and via the ingestion of contaminated deposited sediments by deposit feeders. Unfortunately, sequential extraction data do not give a direct measure of bioavailability to organisms. The amount of a radionuclide available for uptake to non-filter-feeding organisms will depend, to some extent, on the fraction of the radionuclide in soluble form, either in the water column or in pore waters. Studies concentrating on these aspects of geochemistry as oppose to sediment phase geochemistry are therefore likely to yield

results more suited to uptake studies. Sequential extraction data for sediments, instead, provide information on the fraction of the radionuclide in the sediment that can act as a reservoir for potential transfer to biota. A new dimension is introduced into the assessment in the sense that geochemical phase association data can be used to predict the fraction of a given radionuclide in the sediment which may be released if environmental conditions (Eh, pH etc.) change. That fraction of the radionuclide which is likely to be «locked away» from biological interaction over long time periods can also be considered by this method.

In the process of conducting the initial part of a marine impact assessment, the modelling of radionuclides can be arbitrarily split into 2 components namely (i) physical (abiotic) transfer processes and (ii) biological transfer through marine food-chains.

## 2. Contaminant Transport models

In order to simulate some of the processes considered above numerical models need to be constructed and employed.

### 2.1 Model structures

A number of modelling approaches have been used to simulate the physical transport of tracers or contaminants in marine environments varying from uniform and instantaneous mixing to 3 D models where the movement of contaminants can be simulated in the vertical and horizontal planes:

- (i) Box models (or O D) are structured in a way that subdivides the marine environment into large areas over which parameters are averaged. Uniform and instantaneous mixing is assumed to occur in each area and transfer at area boundaries is calculated depending upon model parameters (e.g. interface cross-section and flow rates). Empirical data sets or other models may be required to provide information, e.g. on hydrodynamic flow field, that can be subsequently used to parameterise the model.
- (ii) 1,2 and 3 D models are differentiated in both time and space thus give a much higher spatial resolution. These models are usually solved using finite difference, finite element or stochastic-based particle tracking methods. Due to much greater computational power and input data set (e.g. highly resolved climatic forcing data) requirements, the models are best suited for shorter simulation periods (months-years).

### 2.2. Common Model components

Several simulated processes are common to many of the models considered below. In particular, any model that will be used to simulate the transport of a contaminant in a marine system will account, in some way, for water movement and the advection and dispersion of the contaminant within the water body. Some models additionally simulate the transport of sediment-bound contaminants. Some of these key processes are considered below.

#### 2.2.1 Hydrodynamic processes

Hydrodynamic processes which determine the characteristics of the water flow, i.e. water levels, pressure, velocity, fluxes, salinity, density and temperature, can be modelled. The continuity equation (e.g. Bird et al., 1960) is used to compute the vertical velocities or the water levels, from the horizontal velocity field, depending on its domain of integration (local cell or water column). Navier-Stokes equations (e.g. Bird et al., 1960)) are used to define the relationship between the velocity and the pressure. When the hydrostatic assumption is made, the pressure is proportional to the water level. Other equations may also be required to simulate additional processes including: the transfer of momentum by advection, the turbulent dispersion, the bed friction, the pressure gradients due to the surface elevations (barotropic mode) and to the density differences (baroclinic mode), the Coriolis force generated by the earth rotation on geophysical flows, etc. Physical processes that influence the hydrodynamics and are generated at the boundaries of the domain by external phenomena (tides, waves, wind, heating and cooling, discharge of a river) or occur within the domain (diffusion and dispersion, bed friction) can also be modelled.

#### 2.2.2 Advection–dispersion

Models have been developed to simulate the advection-dispersion of contaminants assuming that concentration changes, etc. have no significant effect on the hydrodynamics. The concentration of the

substance in time and space is computed using advection-diffusion equations. In addition to the transport of the substance by advection and turbulent dispersion, the equation can include buoyancy, decay terms, or source-sink terms to represent the adsorption-desorption on cohesive sediments. Various kinds of substances can be considered in advection-dispersion models: dissolved or particulate matter, conservative or decaying tracers interacting or not with other substances (sediment, salt, etc.), substances having different weight or density such as oil slicks. The salinity can be considered as a purely conservative tracer, (i.e. an element that reacts negligibly with the particulate phase and the concentration for which follows a linear mixing line). Therefore it can be used for the calibration of purely advective and diffusive effects (i.e. effects that exist in the absence of sediment interactions etc.). The majority of radionuclides discharged into the aquatic environment are metallic elements. Hence, processes involved in heavy metals modelling may also be used for radioactive discharge modelling. Distribution coefficients ( $K_{d,s}$ ; defined as the ratio between the solid and solution phase concentrations) are often used to describe the equilibrium balance between dissolved and particulate phases (normally for the pelagic environment) assuming that exchanges of radionuclides between particulate phases and water are wholly reversible.

### 2.2.3 Contaminated Sediment transport models

Modelling dispersion may require the modelling of the transport of radionuclides associated with mineral sediments. A range of sediment transport models have been developed. The most suitable models are box or 2D horizontal models, that include a wide range of processes (e.g. hydrodynamics, advection-dispersion process, sediment dynamics and biological activity). Sediment dynamics modelling will need to consider the type of sediment (i.e. non-cohesive sands and gravels and cohesive silts and clays). Advection-dispersion equations that include vertical settling (Stokes' Law and empirical laws of settling velocity) and erosion terms represent the transport of sediments by the bed load (rolling on the bottom), saltation (transport of sediments by bouncing along the surface) or suspended load (in suspension in the water column). More complex formulations (Mehta *et al.* 1982; and Hayter, 1986) have been developed to take into account bioturbation of consolidated bed sediments.

## 2.3 Examples of transfer models

Numerous contamination transport models have been developed for use in European marine areas (for estuarine, coastal and open sea environments). The models more widely used in impact assessments, include:

- The 2D model **VERSE** is capable of simulating the hydrodynamics, sediment dynamics, radionuclide and trace metal dispersion in partially mixed estuaries (Gleizon, 2002).
- **DIVAST** is a 2-D hydrodynamic, solute and sediment transport model for estuarine and coastal waters.
- **ECoS** is a modelling environment as oppose to a true model. Templates guide the user through the set up and running of an estuarine model; however, these can be fully adapted according to the users requirements (reference) The model was initially designed to concentrate on biogeochemical reactions with limited representation of physical transport properties.
- **MIKE21** (DHI, 2001) is a modular 2D modelling system for free surface flows. It is widely used for hydraulic modelling in estuaries, coastal waters, seas and also lakes.
- The Delft Hydraulics pilot model is a two dimensional depth-integrated model of the North Sea (Postma *et al.*, 1987) developed to study the long-term impact of pollution from river discharges.

- The **BSH model**, developed to study the dispersion of  $^{99}\text{Tc}$  from Cap de la Hague, in the English Channel and the North Sea (Schönfeld, 1995), is a 3-D baroclinic circulation and Lagrangian dispersion model.
- The **GHER model** is a 3D model used to determine the residual dispersion of  $^{137}\text{Cs}$  on the European continental shelf seas (Djenidi *et al*, 1987).
- The **IFREMER model** is a 2D depth-integrated Lagrangian model of the English Channel (Salomon *et al*, 1987) and has been used to determine the residual dispersion of  $^{125}\text{Sb}$  from Cap de la Hague, in the Golfe Breton-Normand.
- **TELEMAC** is a modelling system for simulating physical processes associated with estuaries, coastal waters and rivers including: steady state flow; tidal, wind or wave-driven hydrodynamics; dispersion of pollutants including heat, transport, erosion and deposition of sand and mud; water quality; and wave dynamics.
- **PLUMES PLUME-RW** is a well-established model developed for studies of pollutant dispersion in estuaries and coastal waters. Both dissolved pollutants, such as bacteria from sewage discharges, and suspended pollutants, such as sediment released during dredging operations, can be simulated.
- **MEAD** is used for the prediction of long-term radionuclide dispersion in the Irish Sea by modelling the variation in annually averaged radionuclide activity concentrations over periods up to 100 years. It includes interaction between bed and suspended sediment. The most recent version also considers ionic exchange processes between the dissolved phase and the bed sediment.
- **The POLCOMS model** has been developed to model the dispersion of  $^{137}\text{Cs}$  in and out of the Irish Sea (Prandle, 1984) covering the European continental shelf seas (Irish Sea, North Sea and English Channel). The model is depth-integrated and includes tides and winds, as well as the effects of horizontal density gradients.
- **POSEIDON** is a box model of the European continental shelf seas. It does not solve the hydrodynamics of the region but determines the path and dispersion of radioactive contaminants.
- **DORIS** is a marine dispersion model for European waters capable of calculating activity concentrations in seawater and marine sediments.
- **CSERAM** is a model for prediction of marine radionuclide transport in both particulate and dissolved phases (Aldridge, 1998). The model attempts to go beyond the traditional box model approach in describing the underlying physical processes in a more realistic way. CSERAM includes a 2-D hydrodynamic description of the tidal and wind-induced flows; a wind-wave model to provide the wave-induced bed stress that controls the behaviour of the suspended and settled sediments; and, a physically-based transport model to simulate the movement of both the dissolved and particle-bound radionuclides.
- **Perianez model (University of Sevilla)**. 3-D finite difference model with particle reaction kinetic.

## 2.4 Examples of transfer models with specific application to the Arctic

Several models have been specifically applied within the Arctic environment including:

- **NAOSIM** (North Atlantic-Arctic Ocean Sea Ice Model ; reference) is a 3-D coupled ice-ocean model covering the Arctic Ocean, the Nordic Seas and the North Atlantic north of  $50^\circ\text{N}$ . The model has been used previously to investigate the circulation of ice and ocean currents in the Arctic Ocean and Nordic seas. It has been applied to simulate: (i) the dispersion of  $^{99}\text{Tc}$  released from the Sellafield reprocessing plant to northern seas (Karcher *et al.*, 2002); and (ii) the

potential spread of radioactivity following a hypothetical release from the Kursk submarine (Gerdes et al., 2001).

- **HAMSOM/VOM** (reference) is a 3-D, baroclinic, coupled ice-ocean circulation model. It is based on non-linear basic equations of motion, invoking the hydrostatic approximation and the equation of continuity, which serves to predict the elevation of the free surface from the divergence of the depth mean transport. The numerical scheme is semi-implicit and the equations are discretised as finite difference equations on an Arakawa C-grid. The circulation model includes an Eulerian transport algorithm for temperature, salinity and passive tracers, based on the advection-diffusion equation. HAMSOM/VOM is coupled to a thermodynamic and dynamic sea ice model, which calculates space and time dependent variations of ice thickness and ice concentration. Sea surface heat fluxes are used to determine the ocean temperature and thermodynamic ice formation. The model has been recently applied in the modelling of contaminant transport in Arctic shelf seas and estuaries (Harms, 1997; Harms *et al.*, 2002).
- The **NRPA marine box model** is an improved version of the compartmental model developed by Nielsen *et al.* (1997). The model is based on the modified approach for box modelling (Iosjpe *et al.*, 2002a), which includes dispersion of radionuclides during time (non-instantaneous mixing in oceanic space).
- **MARINA II box model** covering Europe and Arctic marine environments.

Compartmental/box modelling has been recommended by the European Commission for radiological assessment (EC, 1995). Reasons for selecting box models in spite of fact that 3-D hydrodynamic modelling can provide more detailed information especially for short time and distance scales include:

- Three-dimensional hydrodynamic models often require complete, site-specific information concerning meteorological conditions over short time intervals. Although these data are available historically they are obviously not available for the future and predictions will therefore contain a high degree of uncertainty. Predictions made using temporally and spatially averaged (input) data, within a box modelling environment, are likely to have less uncertainty associated with them in assessments where prognoses for long time scales are of interest.
- The most sensitive parameter affecting doses assessments are concentration factors (CF) used to predict radionuclide levels in biota (Iosjpe & Borghuis 2000). The uncertainty associated with CF outweighs the advantages offered by 3-D hydrodynamic models.
- The high spatial resolution associated with 3-D hydrodynamic models means that a similar level of resolution describing the movement of marine biota in oceanic space is required so that points of coincidence between contaminant plumes and organisms can be identified and biological uptake determined. These types of data are rarely available.

However, 3-D hydrodynamic models can be used to improve the oceanic space structure and water fluxes in box models (Karcher & Harms, 2000).

## 2.5 Prediction of radionuclide contamination in Arctic seas using the NRPA marine box model

The NRPA marine box model, which is routinely used by NRPA for marine dose estimates for Man, has been selected for the prediction of Arctic sea radionuclide contamination in the project “EPIC”. Whilst in part this is because the model is available for us to use, it also has a number of advantages for environmental impact assessment in Arctic environments.

### 2.5.1 Detailed description of the NRPA marine box model

Equations of the transfer of radionuclides between boxes are of the form:

$$\frac{dA_i}{dt} = \sum_{j=1}^n k_{ji} A_j - \sum_{j=1}^n k_{ij} A_i \gamma(t \geq T_j) - k_i A_i + Q_i, \quad t \geq T_i$$

$$A_i = 0, \quad t < T_i$$
(2.1)

where  $k_{ii}=0$  for all  $i$ ,  $A_i$  and  $A_j$  are activities (Bq) at time  $t$  in boxes  $i$  and  $j$ ;  $k_{ij}$  and  $k_{ji}$  are rates of transfer ( $y^{-1}$ ) between boxes  $i$  and  $j$ ;  $k_i$  is an effective rate of transfer of activity ( $y^{-1}$ ) from box  $i$  taking into account loss of material from the compartment without transfer to another, for example radioactive decay;  $Q_i$  is a source of input into box  $i$  ( $Bq y^{-1}$ );  $n$  is the number of boxes in the system.  $T_i$  is the time of availability for box  $i$  (the first time when box  $i$  is open for dispersion of radionuclides) and  $\gamma$  is a unit function such that when  $t \geq T_i$   $\gamma = 1$  and when  $t < T_i$ ,  $\gamma = 0$ . The times of availability  $T_i$  are calculated as a minimized sum of the weights for all paths  $\mu_o(v_o, \dots, v_i)$  from the initial box ( $v_o$ ) with discharge of radionuclides to the box  $i$  on the oriented graph  $G=(V, E)$  with a set  $V$  of nodes  $v_j$  correspondent to boxes and a set  $E$  of arcs  $e_{jk}$  correspondent to the transfer possibility between the boxes  $j$  and  $k$ . Every arc  $e_{jk}$  has a weight  $w_{jk}$  which is defined as the time required before the transfer of radionuclides from box  $j$  to box  $k$  can begin (without any way through other boxes).  $M_i$  is a set of feasible paths from the initial box ( $v_o$ ) to the box  $i$  ( $v_i$ ).

Therefore:

$$T_i = \min_{\mu_m(v_o, v_i) \in M_i} \sum_{j,k} w_{jk}$$
(2.2)

Figure 2.1 shows the structure of the model compartments for the Arctic Ocean, the Nordic Seas and the North Atlantic. Each box has surface, mid-depth and deep waters layers (Figure 2.2) based on a knowledge of water fluxes (Karcher & Harms, 2000) and site-specific information. The volume of the water layers in each box has been calculated using detailed bathymetry (IBCAO, 2001 ETOPO5, 2002). The model includes the processes of advection of radioactivity between compartments, sedimentation, diffusivity of radioactivity through the pore water, resuspension, mixing due to bioturbation and a burial process of activity in deep sediment. Radioactive decay is included in all compartments. The contamination of biota is calculated from the radionuclide concentrations in filtered seawater (often nominally defined by 0.45 micron or 1 micron filters) in the different water regions.



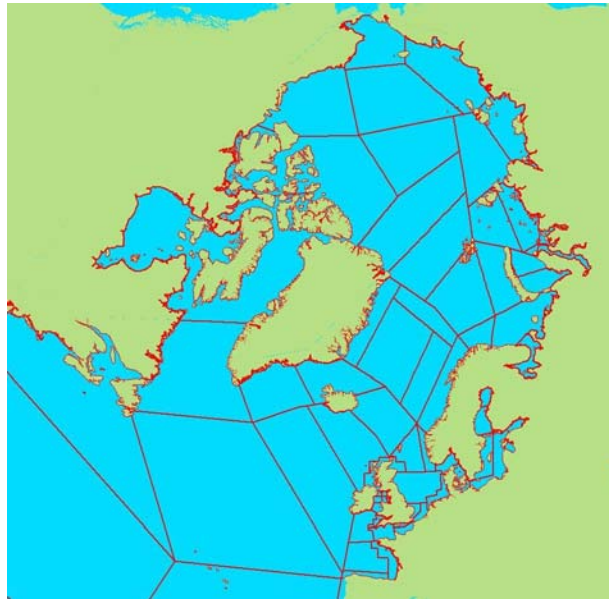


Figure 2.1 The structure of the surface water boxes.

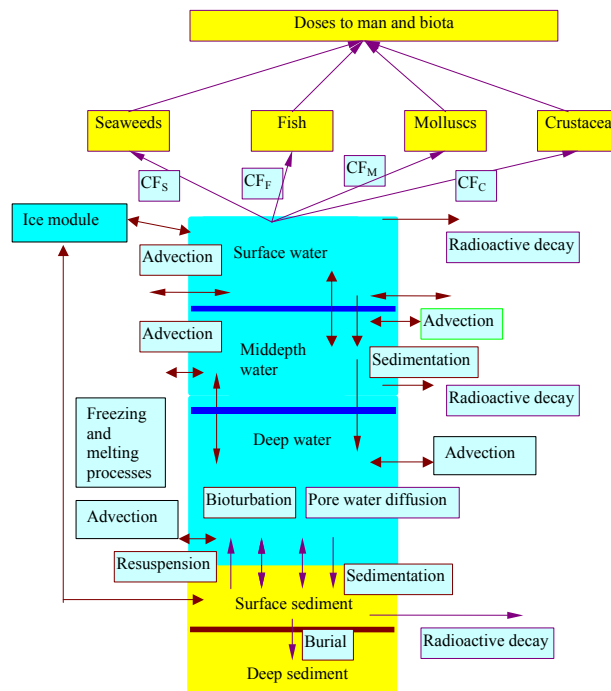


Figure 2.2 Schematic structure of the processes modelled in each box of the NRPA marine model.

### 2.5.2 Adaptation to the Arctic – the ice module

The model can predict radionuclide exchanges between water and ice phases (Iosjpe, 2002a; Iosjpe,2002b). Transfer of radioactivity,  $A_i$  (Bq), from the liquid phase and the suspended sediment in the water column of the water box  $i$  with sediment distribution coefficient  $K_d$  and suspended sediment load  $SSL_i$  from the marine boxes of the NRPA marine box model to the ice box is described as:

$$\lambda_L = A_i \frac{R^{(iw)} \cdot f_i^{(f)}}{1 + K_d \cdot SSL_i} \quad (2.3)$$

$$\lambda_S = A_i \frac{K_d \cdot SL_i^{(I)}}{1 + K_d \cdot SSL_i} \cdot \varphi_i^{(ss)} \cdot f_i^{(f)}$$

where  $\lambda_L$  = transfer of radioactivity from the liquid phase of the water column of box I to the ice box;  $\lambda_S$  = transfer of radioactivity from the suspended particulate phase of the water column of box I to the ice box.  $R^{(iw)}$  is the ice-water transfer factor, corresponding to the fraction of radioactivity, which is transferred from the liquid phase of the sea water box to the ice box during the freezing process (dimensionless),  $f_i^{(f)}$  is a freezing rate for the ice box I ( $m^3 y^{-1}$ ),  $SL_i^{(I)}$  is the total ice sediment load for the ice box I ( $t m^{-3}$ ),  $\varphi_i^{(ss)}$  is a fraction of suspended sediment in water column of the water box  $i$  in sediment of the ice box ( $\sum_i \varphi_i = 1$ ) (dimensionless).

The transfer of radioactivity from the sediment box  $i$  to the ice box is described as

$$A_i^{(s)} \frac{K_d \cdot SL_i^{(I)}}{\omega + (1 - \omega) \cdot \rho_s K_d} \cdot \varphi_i^{(s)} \cdot f_i^{(f)}, \quad (2.4)$$

where  $A_i^{(s)}$  is activity (Bq) in the sediment box  $i$ ,  $\varphi_i^{(s)}$  is a fraction of sediment from the sediment box  $i$  in sediment of the ice box,  $\omega$  is a the porosity and  $\rho_s$  is a sediment density from the marine part of the NRPA marine box model.

Expressions (2.3 and 2.4) are written assuming that transfer of radioactivity varies as a linear function of the freezing rate. The transfer of radioactivity from the ice box  $i$  to the ice box  $j$  is described as:

$$A_i^{(I)} \cdot t_{ij}^{(r)}, \quad (2.5)$$

where  $A_i^{(I)}$  is radioactivity in the ice box  $i$  and  $t_{ij}^{(r)}$  is the ice flux from the ice box  $i$  to the ice box  $j$ .

The transfer of radioactivity through melting process from the ice box  $i$  to the box  $j$  of the marine box model, which underlies the ice box  $i$  is described as

$$A_i^{(I)} \cdot (f_j^{(m)} - \sum_k t_{ik}^{(r)}). \quad (2.6)$$

where  $f_j^{(m)}$  is a melting rate for the ice box  $i$ .

Parameters  $f_i^{(f)}$ ,  $f_i^{(m)}$  and  $t_{ij}^{(r)}$  must satisfy the expression

$$f_i^{(f)} - f_i^{(m)} - \sum_k t_{ik}^{(r)} = 0 \quad (2.7)$$

for each ice box  $i$ .

### 2.5.3 Example simulations

Applications of the NRPA box model in the context of an environmental impact assessment are illustrated in figures 2.3 – 2.5. All simulations have been made for a 1 TBq discharges of radionuclides into Obskaya Guba (the Ob Estuary of the Kara Sea). Dynamic concentrations of  $^{137}\text{Cs}$  and  $^{239}\text{Pu}$  are

shown for cod, crab (muscles) Greenland seal for the Obskaya Guba and the Barents Sea in Figures 2.3 and 2.4. Calculations relating to the uptake by and transfer to biota are based on generic concentrations factors derived specifically for Arctic marine reference organisms.

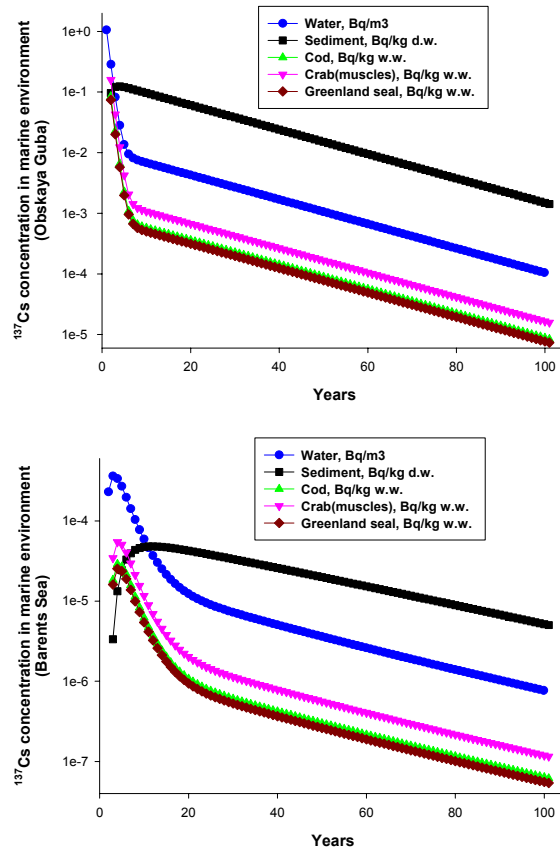


Figure 2.3  $^{137}\text{Cs}$  dynamic concentration in marine environment.

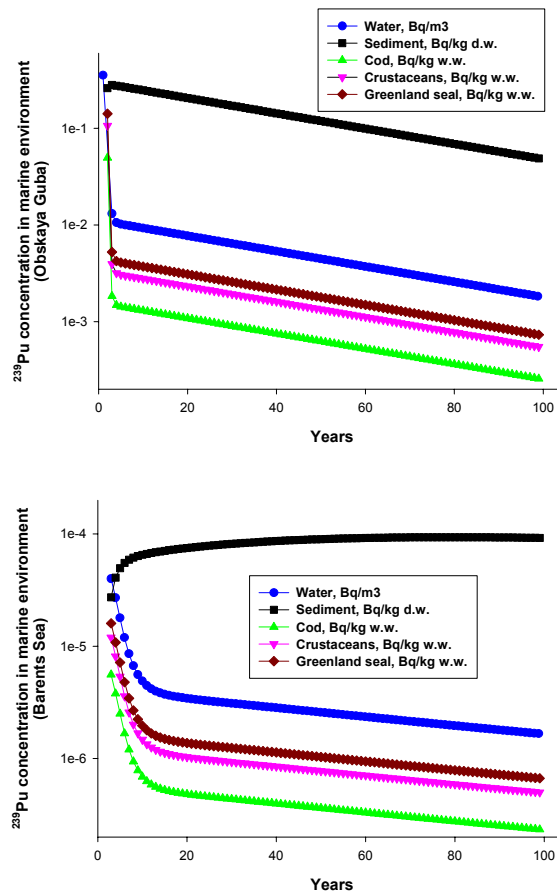


Figure 2.4  $^{239}\text{Pu}$  dynamic concentration in marine environment.

The influence of innovative components of the present box modelling approach is illustrated in Figure 2.5. The simulation corresponds to the dispersion of 1 TBq of  $^{241}\text{Am}$  discharged into Obskaya Guba and accounts for ice transport of radionuclides from the Kara Sea to the Greenland Sea through the Central Arctic Basin. The dynamics of  $^{241}\text{Am}$  concentrations in lobsters living in the Greenland Sea is calculated with a generic Arctic concentration factor. Figure 2.5 clearly demonstrates that ice transport of radionuclides can be a significant factor for some scenarios and radionuclides. It has been shown that the influence of ice transport increases with increasing  $K_d$  values for radionuclides (Iosjpe, 2002b; Iosjpe *et al.*, 2002).

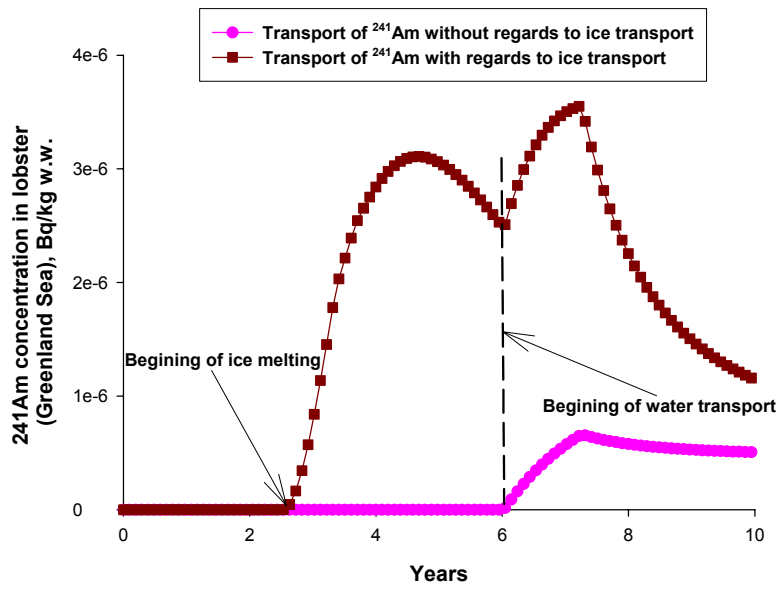


Figure 2.5  $^{241}\text{Am}$  concentration dynamic with and without ice transport of radionuclide.

## 3. Transfer to biota under equilibrium conditions

### 3.1 Introduction

Concentration factors<sup>1</sup> have been widely used in modelling the transfer of radionuclides from the water column to biota and therefore numerous reviews and summaries of the available literature have been made (Harrison, 1986; Gomez *et al.* 1991). Probably the most widely-used concentration factor values, in the fulfilment of human dose assessments, are those reported by the International Atomic Energy Agency in Techdoc. 247 (IAEA, 1985) and the updated version of this document. Selected data from these publications corresponding to the radionuclides chosen for further analysis within the FASSET project (with the exception of <sup>40</sup>K), are shown in Table 3.1.

*Table 3.1 Concentration factors for generic marine organisms (updated version of IAEA Techdoc. 247)*

Element	Phytoplankton	Macroalgae	Zooplankton	Mollusca*	Crustaceans	Fish
Cs	2 x 10 <sup>1</sup>	5x10 <sup>1</sup>	4x10 <sup>1</sup>	6x10 <sup>1</sup>	5x10 <sup>1</sup>	1x10 <sup>2</sup>
Tc	<b>4 x 10<sup>0</sup></b>	<b>3 x 10<sup>4</sup></b>	1 x 10 <sup>-2</sup>	<b>5 x 10<sup>2</sup></b>	1 x 10 <sup>-3</sup>	<b>8 x 10<sup>1</sup></b>
Sr	<b>1 x 10<sup>0</sup></b>	<b>1 x 10<sup>1</sup></b>	<b>2 x 10<sup>0</sup></b>	<b>1 x 10<sup>1</sup></b>	<b>5 x 10<sup>0</sup></b>	<b>3 x 10<sup>0</sup></b>
U	2 x 10 <sup>1</sup>	1 x 10 <sup>-2</sup>	<b>3 x 10<sup>1</sup></b>	3 x 10 <sup>1</sup>	1 x 10 <sup>1</sup>	1 x 10 <sup>0</sup>
Th	<b>4 x 10<sup>5</sup></b>	2 x 10 <sup>-2</sup>	1 x 10 <sup>-4</sup>	1 x 10 <sup>-3</sup>	1 x 10 <sup>-3</sup>	6 x 10 <sup>-2</sup>
Pu	<b>2 x 10<sup>5</sup></b>	<b>4 x 10<sup>3</sup></b>	<b>4 x 10<sup>3</sup></b>	3 x 10 <sup>3</sup>	<b>2 x 10<sup>2</sup></b>	<b>1 x 10<sup>2</sup></b>
Am	2 x 10 <sup>5</sup>	8 x 10 <sup>3</sup>	<b>4 x 10<sup>3</sup></b>	<b>1 x 10<sup>3</sup></b>	<b>4 x 10<sup>2</sup></b>	<b>1 x 10<sup>2</sup></b>
Cm	<b>2 x 10<sup>5</sup></b>	<b>5 x 10<sup>3</sup></b>	<i>4 x 10<sup>3</sup></i>	<b>1 x 10<sup>3</sup></b>	<i>4 x 10<sup>2</sup></i>	<b>1 x 10<sup>2</sup></b>
Np	1 x 10 <sup>-2</sup>	5 x 10 <sup>1</sup>	<i>4 x 10<sup>2</sup></i>	4 x 10 <sup>-2</sup>	<i>1 x 10<sup>-2</sup></i>	<b>1 x 10<sup>0</sup></b>
Ra	2 x 10 <sup>3</sup>	1 x 10 <sup>-2</sup>	1 x 10 <sup>-2</sup>	<b>1x10<sup>-2</sup></b>	1 x 10 <sup>-2</sup>	<b>1 x 10<sup>-2</sup></b>
Pb	<b>1 x 10<sup>-5</sup></b>	1 x 10 <sup>-3</sup>	1 x 10 <sup>-3</sup>	<b>5 x 10<sup>4</sup></b>	<b>9 x 10<sup>4</sup></b>	2 x 10 <sup>-2</sup>
Po	<b>7 x 10<sup>4</sup></b>	1 x 10 <sup>-3</sup>	3 x 10 <sup>-4</sup>	<b>2 x 10<sup>4</sup></b>	<b>2 x 10<sup>4</sup></b>	2 x 10 <sup>-3</sup>
C	9 x 10 <sup>3</sup>	1 x 10 <sup>4</sup>	2 x 10 <sup>4</sup>	2 x 10 <sup>4</sup>	2 x 10 <sup>4</sup>	2 x 10 <sup>4</sup>
H	1 x 10 <sup>0</sup>	1 x 10 <sup>0</sup>	1 x 10 <sup>0</sup>	1 x 10 <sup>0</sup>	1 x 10 <sup>0</sup>	1 x 10 <sup>0</sup>
Nb	1 x 10 <sup>3</sup>	3 x 10 <sup>3</sup>	<i>2 x 10<sup>7</sup></i>	1 x 10 <sup>3</sup>	2 x 10 <sup>2</sup>	3 x 10 <sup>1</sup>
Ni	3 x 10 <sup>3</sup>	2 x 10 <sup>3</sup>	1 x 10 <sup>-3</sup>	2 x 10 <sup>3</sup>	1 x 10 <sup>3</sup>	1 x 10 <sup>3</sup>
Ru	2 x 10 <sup>5</sup>	2 x 10 <sup>3</sup>	3 x 10 <sup>-4</sup>	<b>5 x 10<sup>2</sup></b>	1 x 10 <sup>-2</sup>	2 x 10 <sup>0</sup>
I	<b>8 x 10<sup>-2</sup></b>	<b>1 x 10<sup>4</sup></b>	3 x 10 <sup>-3</sup>	1 x 10 <sup>1</sup>	<b>3 x 10<sup>0</sup></b>	<b>9 x 10<sup>0</sup></b>
Cl	1 x 10 <sup>0</sup>	5 x 10 <sup>-2</sup>	1 x 10 <sup>0</sup>	5 x 10 <sup>-2</sup>	<b>6 x 10<sup>-2</sup></b>	<b>6 x 10<sup>-2</sup></b>

Values in bold indicate those that have been updated from IAEA (1985).

Italicized = best estimates

\*excluding cephalopods

The CF approach has the advantage of being simple and provides the assessor with a large and easily-accessible data-base. It therefore provides a useful starting point for our assessment of transfer and uptake of radionuclides within any marine environmental impact assessment.

The updated version of IAEA Techdoc. 247 also provides information on uptake to marine mammals for selected radionuclides/elements as shown in Table 3.2.

<sup>1</sup> The concentration factor (CF) is usually defined as the ratio of the concentration of the radionuclide in the organism or tissue to that in the ambient seawater.

*Table 3.2 Data on marine mammals from the updated version of IAEA Techdoc. 247*

Element	Pinniped muscle	Pinniped liver	Polar Bear muscle	Polar Bear liver	Cetacean muscle	Cetacean Liver
Cs	$4 \times 10^2$	$3 \times 10^2$	$1 \times 10^2$	n.a.	$3 \times 10^2$	n.a.
Ni	n.a.	n.a.	n.a.	n.a.	$< 2 \times 10^3$	n.a.
Pb	$3 \times 10^3$	$1 \times 10^5$	n.a.	n.a.	$4 \times 10^4$	$6 \times 10^4$
Pu	n.a.	$8 \times 10^0$	$7 \times 10^1$	n.a.	n.a.	$3 \times 10^0$

n.a. – not available

### 3.2 Applicability of CF data to transfer–uptake assessments for non–human biota.

Although the generic organism groups considered in IAEA (1985) and the updated version of this document are similar, and in some cases identical, to those selected as reference organisms within FASSET (Table 3.3), the applicability of these data to the present work is partly limited.

*Table 3.3 (candidate) reference organisms selected in FASSET (see Strand et al., 2001)*

<b>Bacteria</b>	<b>Crustacean</b>	<b>Mammal</b>
<b>Worm</b>	<b>Bivalve Mollusc</b>	<b>Wading bird</b>
<b>Vascular plant</b>	<b>Benthic fish</b>	<b>Phytoplankton</b>
<b>Macroalgae</b>	<b>Pelagic Fish</b>	<b>Zooplankton</b>

In view of the fact that the intended use of CF data would be in human dose assessments, the approach adopted in IAEA (1985) and the updated version of this document involved the collation of data for organism forming parts of food-chains leading to man, i.e. edible plants and animals. Furthermore, to the extent possible with the available data, the information was reported for the edible body parts of these edible organisms. Clearly, a question of data compatibility exists here. Within marine environmental impact assessments, non-edible organisms forming parts of food-chains that have no connection with man should be given equal consideration to those dealt with in human dose-assessment. It is also clearly of importance to consider not only those parts of an organism eaten by man but also those body parts that might be of interest from a dosimetric or dose-effects perspective for the organism per se. Such organs/body parts might include, where relevant, the hepatic system (where high accumulation of heavy metal contaminants can occur) and gonads (important from the perspective of fertility).

In view of these limitations, a data collation exercise was conducted at the NRPA in order to derive information that would be of use in an environmental impact assessment. The data collated in the following section of the report are intended to provide a substantial supplement to the more generic values provided in IAEA (the updated version of this IAEA Techdoc. 247).

### 3.3 Concentration factors

The concentration factor method assumes that the organism is in biochemical equilibrium with its surroundings. The time required for equilibrium to be attained depends on the half-life of the radionuclide and the biological half-life of the element in the organism (Till and Meyer, 1983). The physicochemical form of the element and its route of entry into the organism are among factors that affect CF value. Radionuclides may exist in different physicochemical forms with a distribution that

varies according to the radionuclide and the features of the ecosystem under consideration. Environmental factors, including temperature, light (in the case of algae), salinity and pH affect the growth and metabolism of organisms, and consequently the uptake of radionuclides (Meinhold *et al.* 1991).

### 3.4 Scope of CF data collation exercise

Owing to time constraints certain limiting search criteria were applied.

- (i) Publications older than 1984 were not included in this report based on the assumption that these data were considered in a comprehensive manner in IAEA (1985). With extended time and resources it would be fruitful to revisit the data compiled in this tech doc, and other historical data-sets, and reconfigure them in a form more suitable for biota impact assessment.
- (ii) The focus of this study has been on concentration factor data pertaining to European marine ecosystems<sup>2</sup> in line with the objectives of the FASSET project.
- (iii) Potassium-40 has not been considered further owing to the fact that a scenario whereby this radionuclide would be released at rates high enough to significantly augment the activity concentrations above those observed under natural conditions could not be envisaged.
- (iv) (benthic) Bacteria were not considered in terms of CFs. External exposure from contaminated sediments will completely dominate the dose rate to these organisms.
- (v) Only data pertaining to radionuclides has been collated. A means of augmenting the data set would involve the collation of CFs derived from the simultaneous measurement of stable isotopes in biota and seawater.

### 3.5 Organization of the collected data

Data compiled from the literature were organized (sorted) into subsets of interest using Excel spreadsheets. These compiled data were first divided geographically into European and non-European subsets and then each divided further methodologically into: Field data and data from laboratory experiments. Data subsets were then analysed statistically to produce ranges, medians and means.

Table 3.4 shows categories of marine organisms (along with their subdivisions) for which data have been found. Subdivisions of the original reference organism categories (Table 3.3) were created to accommodate reported differences in uptake between orders or families of organism under each category. In the case of crustaceans, reported differences in the uptake of Tc-99 prompted the inclusion of subdivisions: lobster and crab. Differences in uptake of naturally-occurring Po-210 prompted the creation of the subdivision: prawn and isopod.

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<sup>2</sup> For definition of European marine system boundaries, FASSET Deliverable 1 (Strand *et al.*, 2001; Appendix 2) can be consulted.



*Table 3.4: Organism categories considered in the present study.*

<b>Categories</b>	<b>Subdivisions</b>
<b>Microalgae</b>	None
<b>Zooplankton</b>	None
<b>Macroalgae</b>	Chlorophyta (Green) Phaeophyta (Brown) Rhodophyta (Red) Unspecified
<b>Molluscs</b>	Bivalve Gastropods Cephalopod
<b>Crustaceans</b>	Prawn Shrimp Crab Lobster Isopod
<b>Fish</b>	None
<b>Mammals</b>	None
<b>Worms</b>	None
<b>Seabirds</b>	None

In the original reference organism list, a differentiation was made between benthic and pelagic fish owing to the fact that animals included under these 2 categories might be expected to be exposed to quite dissimilar external dose-rates. In contrast to this, there was no strong evidence to suggest that pelagic and benthic fish would exhibit significantly different CFs and therefore the combination of these 2 groups into the single category “fish” (within a single Excel spreadsheet) was considered to be sensible. It should be noted that the nature of the data-base allows these 2 categories to be differentiated if deemed necessary.

The following outline presents the general compilation scheme used in organizing the collected data.

### **3.6 Categorisation of reported studies (Excel spreadsheets)**

CF values are dependent on a multitude of biological and environmental parameters and the number of these parameters varies widely from study to study. In the present work, an attempt has been made to choose parameters, which are generally addressed in most publications. The following factors were registered, if available, for both field and experimental studies:

- Organism category
- Tissue or organ analysed
- Habitat
- Sampling location
- Number of samples
- Time of measurement
- Location of measurement
- Type of water: filtered or unfiltered

The inclusion of the last entry is due to the fact that the type of water, filtered or unfiltered, used in studies can be a major source of variance, particularly for coastal waters. In addition to these categories, CFs reported from laboratory experiments also include:

- Time of exposure in days
- Exposure concentration
- Biological half-life, if available
- Temperature condition

The last column in the database is entitled comments and contains noted special conditions such as:

- Data reported as a mean or as a range rather than as individual observations
- Data estimated rather than directly measured
- Any special remarks, insights, conclusions, warnings, etc. cited
- Equilibrium condition, whenever information was available, particularly for laboratory data

The tissue/organ categories used in cataloguing the data in this study are listed below. The selection was based on (I) data availability (II) a consideration of where radionuclides might accumulate (III) organs that will be of interest from a dosimetric effects perspective (e. g. gonads):

1. Crustaceans and bivalve: Whole, soft tissues/flesh, shell, hepatopancreas, gill, and other
2. Gastropods: whole, soft tissue/flesh, shell, digestive gland (viscera)
3. Cephalopods: whole, muscle, hepatopancreas
4. Fish: whole, muscle/flesh, bone, liver, gonads, and other.

The excel spreadsheets are not included as part of this report – they are available on request to the authors. These data have been used in the production of summary tables as described below.

### 3.7 General remarks

Data collected here have been extracted from different studies. The amount of information/details reported in each study differs according to the objective(s) of that study. Consequently, in order to reduce this diversity and, at the same time, produce a reasonable database, the following conventions were adopted:

- For data reported as a range, the mean of the reported upper and lower bounds was given. In these cases the number of observations was chosen to be two (the lowest possible number).
- For data reported as smaller/larger than, the recorded limit was chosen.
- Seawater was used by default whenever the type of water was not mentioned.
- The number of observations was set equal to one whenever information about it was not provided.
- Whole body was used by default whenever the tissue or organ was not clearly specified.
- Concentration expressed as dry weights, e.g. Masson *et al.* (1995), were converted to wet weight concentration by multiplying the given value by 0.2 as recommended by IAEA, (1985).

### 3.8 Table of summaries

In order to analyse the compiled data, CF values have been pooled together despite their inevitable diversity. In an attempt to reflect (different aspects of the collected data) this variety, three different averages have been calculated: Median, arithmetic and weighted mean. These along with other parameters are presented in Tables 3.6 to 3.19. The following section presents explanatory information associated with the calculated averages as well as other table parameters.

## N

Total number of reported data that have been used to calculate means and medians. This value does not reflect the number of observations in the studies considered.

## n

This parameter represents total number of observations. Unfortunately this value was among those least reported. Its deduction was not straightforward and often use was made of one or more of the conventions listed above. This value has been used to calculate the weighted means,  $M_{\text{weight}}^*$ .

## Weighted mean

Using N to calculate medians and means can mask the scale of different investigations. In order to overcome this shortcoming the weighted mean was calculated, whenever possible.

## Areas

As indicated earlier, the objective of this review was to collect concentration factor data for European marine environments. In the column entitled 'areas' code numbers have been used to represent the areas where the corresponding investigation has been undertaken. Although the Baltic Sea has been categorized as brackish waters by FASSET, some data pertaining to this area has been included in the present study. Numbers stated correspond to the areas:

1. Black Sea
2. Mediterranean Sea (including Adriatic Sea)
3. North-east Atlantic
4. English Channel
5. Irish Sea
6. North Sea
7. Norwegian Sea
8. Barents Sea
9. Greenland Sea
10. Kattegat and Skagerrak
11. Baltic Sea

## 3.9 Paucity of data

Table 3.5 is an attempt to illustrate the scarcity of data. The grey boxes represent the presence of 1 or more data entries and the blank boxes the absence of data for the given radionuclide-biota intersect. The availability of data indicates that some form of CF can be derived for use in an environmental impact assessment. However, the limitations on the application of the associated CF are not specified here. Such limitations may be imposed by the extrapolation of laboratory derived data to field conditions or on a lack of information for specific body parts or organs that may be of interest/concern from a dosimetric perspective.

During the collation exercise conducted by the NRPA, little information was found for radionuclides such as I, Ru, Ra, Np, Cm and U. No data were found for Th, C, H, Nb, Ni and Cl. Nor have data for vascular plants been found. Marine birds, (polychaete) worms and mammals are particularly poorly characterised using CF datasets.

*Table 3.5 Availability of data in the present study and the updated version of Technical Report Series 247 (IAEA, 1985).*

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\*  $M_{\text{weight}} = \sum ((n_i * (CF)_i) / n)$ ; where  $n = \sum n_i$

Elem.	Mac. Al	Mollusc	Crusta.	Fish	Zoopl.	Phytopl.	Worm	Mamm.	Bird
Cs									
Tc									
Sr									
U									
Th									
Pu									
Am									
Cm									
Np									
Ra									
Pb									
Po									
C									
H									
Nb									
Ni									
Ru									
I									
Cl									

a Includes both benthic and pelagic fish as reported in the main body of text.

Due to the interplay of many factors, both biological and environmental, the variance of CFs is great within and between studies. Sazykina (1998) found that the values of Cs-137 CF for fish (Barents Sea, cod) were not constant, but gradually changed from  $28 \pm 5$  in 1979 up to  $182 \pm 48$  in 1992. This study clearly illustrates what is usually a major source of variation in many studies; lack of equilibrium within the period of observations. Consequently, caution must be used when an organism contamination is predicted by the radionuclide ‘concentration factor’ approach.

### 3.10 Points for discussion

A comparison between field data and those derived from laboratory studies shows that laboratory measurements tend to yield lower CF values than field measurements. This discrepancy can be the outcome of an inevitable problem that may face all experimentalists, namely, the difficulty of designing laboratory experiments that simulate real-world conditions.

According to data presented in Table 3.6, seabirds are prone to comparatively high Cs accumulation. This has been explained by the fact that the birds come into contact with seawater primarily for catching prey and cannot deplete themselves of accumulated contaminants through desorption in the way that other marine organisms can (Fisher *et al.*, 1999).

Brown seaweeds have historically been used as a biological indicator of Tc-99 (Aarkrog *et al.* 1986 & 1987). Hurtgen *et al.* (1988) showed that, generally, apical fragments of *Fucus spiralis* possess a lower content of Tc-99 than the middle and basal ones. This indicated that the older parts of this brown alga accumulate more Tc than younger ones. Crustaceans display a wide range of CF values of Tc-99 with crab at the lower end and lobster at the higher end of this range. The highest CF value belongs to the lobster’s green gland, which has exhibited a CF as high as 65000 in the Irish Sea (Busby *et al.*, 1997).

As it is clear from Table 3.8, the distribution of Po-210 is highly non-uniform with highest accumulations occurring in hepatic and digestive organs. Mussels display a higher affinity for Po-210

relative to winkles or prawns. This can be explained by considering their respective ecological niches and feeding habits (McDonald *et al.* 1993).

Table 3.6 Concentration factors for Cs-137 in European marine organisms obtained from field studies

Organism/Tissue	N	Range	Median	Mean	n	M <sub>weight.</sub>	Areas	References	Comments
<b>Crustaceans</b>	1	52 -	-	-	5	-	8	4	
<b>Fish</b>	15	15 - 189	51.5	72	72	65	1,2,5	11,28,32	
	14	28 - 182	100	108.1	196	83.7	1,6,8	1,3,4	
<b>Mammals</b>	1	13 - 70	42	42	2	-	8	4	
	4	100 - 600	350	350	25	366	3,5,6	11	
<b>Seabird</b>	1	414 -	-	-	7	-	8	4	
	1	530 -	-	-	5	-	8	4	
<b>Molluscs</b>	2	16 - 24	20	20	3	21.3	1,5	15,32	
	2	6 - 15	10.5	10.5	5	-	5	15	
<b>Gastrop.</b>	8	18 - 134	37.5	51.6	26	-	5,6	26	
	2	4 - 6	5	5	5	-	5	15	
	2	20 - 55	37.5	37.5	5	-	5	15	
	5	5 - 35	20	18.4	8	21.2	1,5	15,32	
	2	2 - 4	3	3	5	-	5	15	
<b>Bivalve</b>	11	11 - 83	43	40	23	52	5,6,8	4,26	
	2	1 - 3	2	2	2	-	5	15	
	2	1 - 4	2.5	2.5	5	-	5	15	
<b>Macroalgae</b>	2	26 - 124	75.2	75.2	13	116.4	2,8	4,37	
	3	35.7 - 80	51.5	55.7	4	54.6	1,2	32,37	
	4	14.3 - 39.5	31.7	29.3	7	31.5	1,2	32,37	
	12	17 - 56	34.5	35.3	26	-	4,5,6	26	
	1	31.5 -	-	-	37	-	2	37	Cs-134
	2	46 - 127	86	86	37	-	2	37	Cs-134
	1	15.2 -	-	-	37	-	2	37	Cs-134

*Table 3.7 Concentration factors for Tc-99 in European marine organisms obtained from field studies*

<b>Organism/Tissue</b>	<b>N</b>	<b>Range</b>	<b>Median</b>	<b>Mean</b>	<b>n</b>	<b>M<sub>avg</sub></b>	<b>Areas</b>	<b>References</b>	<b>Comments</b>
<b>Fish</b>					49		5	25	
Edible parts	1	12	-						
Whole	1	486	-				7	8	
<b>Molluscs</b>					27	606	5	10,25	
Soft tissue	7	150	- 970	390					
Brown	5	12200	- 36000	30000	6	25500	4,6,7,10	8,31,33	
<b>Macroalgae</b>									
Apical fragment	1	4000	- 20000	12000			6	33	Brown algae
Middle fragment	1	10000	- 51000	30500			6	33	Brown algae
Basal fragment	1	9000	- 46000	27500			6	33	Brown algae
<b>Crustaceans</b>									
Whole	1	386	-				7	8	
<b>Shrimp/prawn</b>									
Edible parts	1	2800	-	91.5			5	25	
Muscle	4	20	-				5	10	
Hepatopancreas	1	160	-				5	10	
Gills	1	70	-				5	10	
<b>Crab</b>									
Muscle	8	40	- 8000	1085			5,7	8,10,25	
Hepatopancreas	2	2300	- 7700	5000			5	10	
Gills	2	1200	- 1600	1400			5	10	
Shell	2	1100	- 1600	1350			5	10	
Green glands	1	65000	-				5	10	

Table 3.8 Concentration factors for **Po-210** in European marine organisms obtained from field studies

Organism/Tissue	N	Range	Median	Mean	n	M <sub>weig.</sub>	Areas	References	Comments
Whole	4	2800 - 7400	4700	4900			11	24	
<b>Phytoplankton</b>									
Macrozoop. Whole	3	12400 - 25400	17000	18300			11	24	>303µm
Mesozoop. Whole	17	8400 - 140800	35000	42000			11	24	>202µm
Natural composition	23	600* - 140800	30000	36500			3,11	23,24	*Jellyfish
Seaweed	13	70 - 2585	890	1000			4,5,6	26	
<b>Macroalgae</b>									
Whole	3	9000 - 23000	17000	16330			11	22	
<b>Worm</b>									
Liver	1	300000	-	-			3	23	*Epipelagic
Gonad	1	60000	-	-			3	23	
Bone	1	30000	-	-			3	23	
Muscle	1	6000	-	-			3	23	
<b>Crustaceans</b>									
Cardiac fore-gut	1	26700	-	-			5	15	
Hepatopancreas	1	22100	-	-			5	15	
Gill	1	3800	-	-			5	15	
Shell	2	1700 - 5000	-	3350			5	15	
Abdom. Muscle	1	300	-	-			5	15	
Whole*	3	24000 - 38000	28000	30000	20	28300	11	22	*+Deca- and Amphipod
Hepatopancreas	1	377000	-	-			11	22	
Alimentary tract	1	24000	-	-			11	22	
Gill	1	19000	-	-			11	22	
Shell	1	14000	-	-			11	22	
Muscle	1	8000	-	-			11	22	
<b>Molluscs</b>									
Whole	9	2410 - 31590	13780	13723			4,5,6	26	
Digestive glands	1	29000	-	-			5	15	
Pallial complex	1	9700	-	-			5	15	
Total soft tissues	1	5500	-	-			5	15	
Muscle	1	1700	-	-			5	15	



**Table 3.8 (cont.)** Concentration factors for **Po-210** in European marine organisms obtained from field studies

<b>Organism/Tissue</b>	<b>N</b>	<b>Range</b>	<b>Median</b>	<b>Mean</b>	<b>n</b>	<b>M<sub>avg</sub></b>	<b>Areas</b>	<b>References</b>	<b>Comments</b>
<b>Molluscs</b>									
Bivalve									
Whole	18	2200 - 74260	17500	25370			5,6,11	22,26	
Soft tissues	34	3600 - 174000	46426	52380	235	58450	5,9,11	15,21,22,27	
Hepatopancreas	1	66000 -					11	22	
Viscera	1	58400 -					5	15	
Alimentary tract	1	35000 -					11	22	
Gills	2	13100 - 27000		20050	5	24220	5,11	15,22	
Shell	8	3000 - 36000	9500	13000	11	10272	11	22	

**Table 3.9** Concentration factors for **Pu-239+240** in European marine organisms obtained from field studies

<b>Organism/Tissue</b>	<b>N</b>	<b>Range</b>	<b>Median</b>	<b>Mean</b>	<b>n</b>	<b>M<sub>weig.</sub></b>	<b>Areas</b>	<b>References</b>	<b>Comments</b>
<b>Fish</b>	1	- 500					8		4
Muscle									
<b>Macroalgae</b>	19	633 - 16000	3467	4637	34	5221	2,4,8	4,35,36,37	
Brown									
Red and Green	3	1037 - 3760	1320	2040	85	1931	2	36,37	
Unspecified	10	1310 - 4950	2450	2693			5,6		26
<b>Molluscs</b>									
<b>Gastropod</b>									
Whole	25	400 - 4600	1500	1865			4,5,6		26,35
Pallial complex	1	15800 -					5		15
Digestive glands	2	7100 - 9700		8400			5		15
Total soft tissues	1	5700 -					5		15
Shell	2	1500 - 1800		1650			5		15
Muscle	2	600 - 1100		850			5		15
<b>Bivalve</b>									
Whole	9	380 - 2240	1480	1233			5,6		15,26
Byssal threads	1	29500 -					5		15
Viscera	1	4800 -					5		15
Total soft tissue	1	1400 -					5		15
Gill	1	1000 -					5		15
Muscle	1	800 -					5		15

*Table 3.10 Concentration factors for Sr-90 from field studies in European marine organisms*

Organism/Tissue	N	Range	Median	Mean	n	M <sub>weig.</sub>	Areas	References	Comments
<b>Macroalgae</b>	1	182	-				8	4	
Brown									
<b>Mammals</b>									
Seal									
Muscle	1	0.4 - 1.2		0.8			8	4	
Liver	1	0.2 - 3		1.6			8	4	
<b>Fish</b>									
Whole	4	36 - 49	44	43			2	28	
Muscle	1	4					8	4	
<b>Crustaceans</b>									
Shrimp									
Shell	1	26					8	34	
Muscle	1	11 - 19		15			8	34	

*Table 3.11 Concentration factors for Pb-210 in European marine organisms obtained from field studies*

Organism/Tissue	N	Range	Median	Mean	n	M <sub>weig.</sub>	Areas	References	Comments
<b>Macroalgae</b>	13	10 - 440	170	183			4,5,6	26	
Seaweed									
<b>Molluscs</b>									
Gastropod									
Whole	10	60 - 6930	715	1180			5,6	26	
Bivalve									
Whole	10	30 - 7360	575	1508			5,6	26	

*Table 3.12 Concentration factors for U-238 in European marine organisms obtained from field studies*

<b>Organism/Tissue</b>	<b>N</b>	<b>Range</b>	<b>Median</b>	<b>Mean</b>	<b>n</b>	<b>M<sub>wet</sub></b>	<b>Areas</b>	<b>References</b>	<b>Comments</b>
<b>Macroalgae</b>									
Seaweed	13	21 - 130	46	50			4,5,6	26	
<b>Molluscs</b>									
Gastropod	9	12 - 82	23	28.4			5,6	26	
Bivalve	10	4 - 83	13	20.2			5,6	26	

*Table 3.13 Concentration factors for Am-241 in European marine organisms obtained from field studies*

<b>Organism/Tissue</b>	<b>N</b>	<b>Range</b>	<b>Median</b>	<b>Mean</b>	<b>n</b>	<b>M<sub>wet</sub></b>	<b>Areas</b>	<b>References</b>	<b>Comments</b>
<b>Macroalgae</b>									
Red	2	1020 - 3760		2400			2	36,37	
Green	1	836 -					2	36,37	
Brown	1	440 -					2	36,37	
<b>Molluscs</b>									
Gastropod	1	600 - 900		700*			5	40,41,42	
Bivalve	1	200 - 1300	700				5	40,41,42	

Table 3.14 Concentration factors for Cm-242, Ru-106 and I-131 in European marine organisms obtained from field studies.

Organism/Tissue	Radionuc.	N	Range	Median	Mean	n	M <sub>weig.</sub>	Areas	References	Comments
Macroalgae	Cm-242	2	5950 - 51700		28800			2	36	
		1	1520 -					2	36	
		1	1320 -					2	36	
Macroalgae	Ru-106	2	1343 - 3854		2600			2	37	
		1	428 -					2	37	
		1	219 -					2	37	
Macroalgae	I-131	2	12690 - 85000		48800			2	37	
		1	921 -					2	37	
		1	418 -					2	37	
Mussel	I-131			96			2	44	<i>Mytilus edulis</i>	

Table 3.15 Concentration factors for Am-241 in European marine organisms obtained from experimental studies

Organism/Tissue	N	Range	Mean	t <sup>a</sup>	C <sup>b</sup>	T <sup>c</sup>	Areas	References	Comments
Macroalgae	1	329 -	-	12	6840	2	6	13	
	1	437 -	-	12	6840	12	6	13	
Molluscs	1	380 -	-	30	?	2	8	14	60% in soft tissues
	2	150 -	200	50	1000	13±2	1	17	Cs-134, Equilibrium
Crustaceans	1	145 -	-	8	1169	13	2	20	
	1	960 -	-	8	1169	13	2	20	
	1	240 -	-	8	1169	13	2	20	
	1	5 -	-	8	1169	13	2	20	
	2	5 -	25	15	1169	13	2	20	
				2.9x10 <sup>5</sup>					

a Time of exposure in days

b Exposure concentration in Bq/l

c Temperature under which the experiment has been undertaken

**Table 3.16** Concentration factors for **Po-210** in European marine organisms obtained from experimental studies

Organism/Tissue	N	Range	Mean	t <sup>a</sup>	C <sup>b</sup>	T <sup>c</sup>	Areas	References	Comments
Crustaceans Shrimp	1	139	-	21	260	14	2	19	Uptake from seawater
	1	24	-	21	260	14	2	19	
	1	316	-	21	260	14	2	19	
	1	272	-	21	260	14	2	19	
Shrimp	1	168	-	14	260	14	2	19	Uptake from seawater and food
	1	54	-	14	260	14	2	19	
	1	958	-	14	260	14	2	19	
	1	303	-	14	260	14	2	19	

**Table 3.17** Concentration factors for **Pb-210** in European marine organisms obtained from experimental studies

Organism/Tissue	N	Range	Mean	t <sup>a</sup>	C <sup>b</sup>	T <sup>c</sup>	Areas	References	Comments
Crustaceans Shrimp	1	682	-	21	260	14	2	19	Uptake from seawater
	1	47	-	21	260	14	2	19	
	1	297	-	21	260	14	2	19	
	1	1738	-	21	260	14	2	19	
Shrimp	1	663	-	14	260	14	2	19	Uptake from seawater and food
	1	32	-	14	260	14	2	19	
	1	928	-	14	260	14	2	19	
	1	1813	-	14	260	14	2	19	

a Time of exposure in days

b Exposure concentration in Bq/l

c Temperature under which the experiment has been undertaken

Table 3.18 Concentration factors for Cs-137 and Cs-134 in European marine organisms obtained from experimental studies

Organism/Tissue	N	Range	Mean	t <sup>a</sup>	C <sup>b</sup>	T <sup>c</sup>	Areas	References	Comments
Macroalgae	1	3.3	-	12	8950	2	6	13	Cs-134
	1	4.6	-	12	8950	12	6	13	Cs-134
Green	1	2	-	18	1000	16	2?	18	
Fish	1	3	-	18	1000	16	2?	18	
Crustaceans									
Isopod	1	22	-	18	1000	16	2?	18	
Molluscs									
Bivalve	1	14	-	30	?	2	8	14	
Whole	2	2.57	- 2.8	50	1000	13±2	1	17	Cs-134, Equilibrium
Phytoplankton	5	≈0	- 40	10	3x10 <sup>5</sup>	12	?	39	Growing cells,
	5	≈0	- 100	6	3x10 <sup>5</sup>	12		39	Non-growing cells

a Time of exposure in days

b Exposure concentration in Bq/l

c Temperature under which the experiment has been undertaken



Table 3.19 Concentration factors for different radionuclides in European marine organisms obtained from experimental studies

Organism/Tissue	Radionucl.	N	Range	Mean	t <sup>a</sup>	C <sup>b</sup>	T <sup>c</sup>	Areas	References	Comments	
Macroalgae	Brown	1	88.5 -		12	7110	2	6	13		
	Green	1	350 -		10	37000	18-21	6	9		
Macroalgae	Red	1	0.1 - 5	2.5	10	37000	18-21	6	9		
	Brown	2	9000 - 12000	10500	10	37000	18-21	6	9		
Worm	Np-239	1	1.5 -		13	2146000	14±1	6	38	Equilibrium	
Molluscs	Bivalve	Soft tissues	1	14 -		13	2146000	14±1	6	38	
		Shell	1	47 -		13	2146000	14±1	6	38	
	Bivalve	Byssus	1		1040					45	Experimental organism was <i>Perna viridis</i> - not native to European waters
		Foot	1		2.4					45	
		Soft tissue	1		6.4					45	
	Bivalve	Shell	1		2.1					45	
		Whole body	1		3.9					46	
	Shell	1		4.7					46		
	Soft parts	1		4.9					46	Experimental organism not native to European waters	
Crustaceans	Crab	Muscle	2	5 - 8	6.5	8	1169	13	2	20	
		Hepatopancreas	1	2 -		8	1169	13	2	20	
		Exoskeleton	1	70 -		8	1169	13	2	20	
		Gills	1	340 -		8	1169	13	2	20	
			I-131	1		4.3					46

a Time of exposure in days

b Exposure concentration in Bq/l

c Temperature under which the experiment has been undertaken

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## 4. Transfer to biota – dynamic modelling approach

### 4.1 Kinetic modelling approach parameterised using allometric relationships

Aside from general matters of applicability, it should also be noted that the CF approach is open to criticism because :

- (i) it provides no information concerning the types of processes/mechanisms in operation during biological uptake,
- (ii) the relationship between a radionuclide water concentration and the radionuclide concentration within (the organs or whole body of) a high trophic-level organism, deriving most of its contaminant load from ingested food, may not be a simple, linear one and
- (iii) the assumption that the system is under equilibrium, a requirement for CFs to be truly applicable, is often invalid,
- (iv) Even if the generic data for the world oceans are employed (Table 3.1), with the limitations on use considered in Section 3.2 having been accepted, the uptake of many radionuclides to certain reference organism types are poorly, if at all, described. A good example can be presented for sea mammals and birds for which data coverage extends only to a handful of radionuclides and where the great preponderance of data exists for  $^{137}\text{Cs}$ .

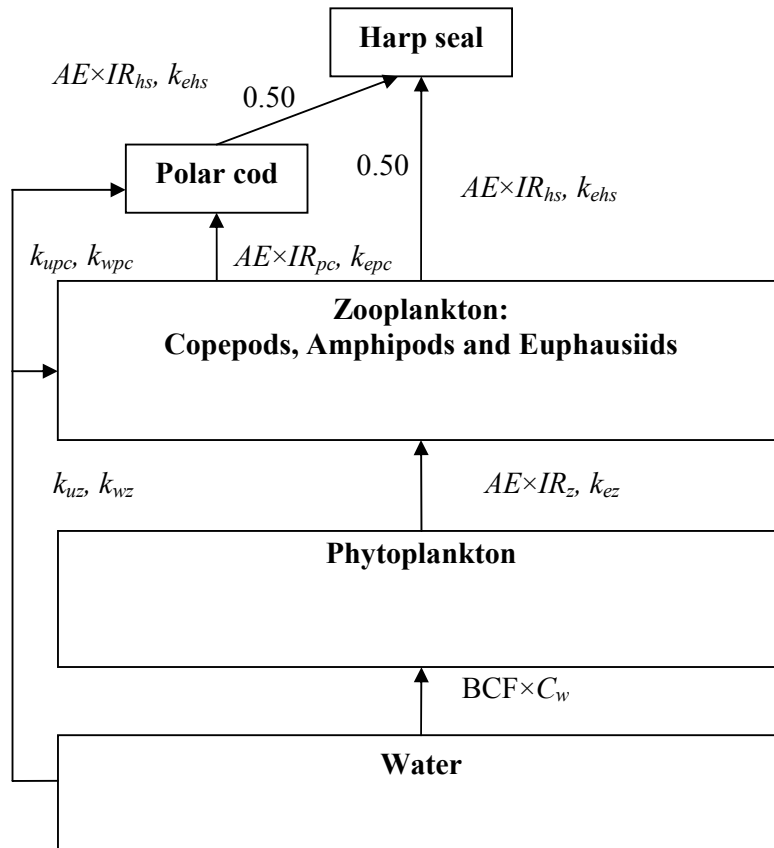
In this chapter other approaches to modeling the transfer of radionuclides in ecological systems will be explored. Biokinetic models may allow more realistic prognoses concerning the dynamic response of an ecological system to be made and allow tentative estimates to be derived concerning equilibrium CFs. Where data are lacking on some of the parameters required for simulation, allometric relationships may provide surrogate values. The allometric approach is based on the observation that metabolic parameters, including basal metabolic rates, ingestion rates, biological half times etc., are proportional to the size of an organism.

#### 4.1.1 Preliminary model structure

In order to demonstrate how this type of model might be employed in the process of filling knowledge gaps, a preliminary version of a food-chain model has been developed to consider the transfer of selected radionuclides ( $^{137}\text{Cs}$  and  $^{239,240}\text{Pu}$ ) to reference organisms in a pelagic foodchain – specifically harp seal, polar cod and zooplankton. The structure of the foodchain is based on information in the open literature (Dommasnes et al., 2001) and is represented in Figure 4.1.

The model, based on earlier work (Thomann, 1981; Landrum et al., 1992; Fisher, 2002) considers uptake via food and water for aquatic organisms, while the excretion/elimination rate is considered to be independent of the uptake route, and the assimilation efficiency is considered to be independent of food type. A further simplification is that the phytoplankton and the zooplankton (trophic levels 1 and 2) are considered as homogeneous groups described by specified parameter values rather than ranges. We also make the simplifying assumption that the growth rate for all organisms is 0. This latter assumption may be a particularly poor one (Thomann, 1981), but the complexity of the weight dynamics for the organisms in question will require more detailed study than can be afforded at the present time.

As an example, the foodchain transfer of a particular radionuclide to harp seal has been conceptualized by the model in Figure 4.1.



**Figure 4.1** Foodchain model for harp seal in the Barents Sea, simplified from Dommasnes *et al.* (2001).

The time-dependent transfer of radionuclides within the foodchain can be described by simple first order differential equations.

**Trophic level 1: Phytoplankton (equilibrium with water concentration):**

$$C_p = BCF \cdot C_w \quad (4.1)$$

where :  $C_p$  = Concentration in phytoplankton (Bq/kg w.w.); BCF (Bioconcentration factor for phytoplankton,  $l \text{ kg}^{-1}$ );  $C_w$  = concentration in water ( $\text{Bq l}^{-1}$ )

The uptake of actinides by phytoplankton cells, under laboratory conditions reach equilibrium with their ambient media with respect to isotope partitioning within a few days (Fisher *et al.*, 1983). This is also true for other actinides including Am, Cf, and Np. This supports (at least partially) our simplifying assumption at the basis of the model, i.e. that equilibrium between seawater and phytoplankton occurs instantaneously.

## Trophic level 2: Zooplankton (uptake via water and food):

$$\frac{dC_z}{dt} = AE_z \cdot IR_z \cdot C_p + k_{uz} \cdot C_w - C_z \cdot k_{ez} \quad (4.2)$$

$AE_z$  = assimilation efficiency (dimensionless) for zooplankton;  $IR_z$  = ingestion rate per unit mass of zooplankton ( $\text{kg d}^{-1}$  per  $\text{kg}$ , w.w.);  $C_p$  = activity concentration in phytoplankton ( $\text{Bq kg}^{-1}$ , w.w.);  $k_{uz}$  = uptake rate of radionuclide to zooplankton directly from water column ( $\text{d}^{-1}$ ),  $C_w$  = activity concentration in water ( $\text{Bq l}^{-1}$ );  $C_z$  = activity concentration in zooplankton ( $\text{Bq kg}^{-1}$ , w.w.);  $k_{ez}$  = excretion rate from zooplankton ( $\text{d}^{-1}$ )

## Trophic level 3: Polar Cod (uptake via water and food):

$$\frac{dC_{pc}}{dt} = AE_{pc} \cdot IR_{pc} \cdot C_z + k_{upc} \cdot C_w - C_{pc} \cdot k_{epc} \quad (4.3)$$

$AE_{pc}$  = assimilation efficiency (dimensionless) for polar cod;  $IR_{pc}$  = ingestion rate per unit mass of polar cod ( $\text{kg d}^{-1}$  per  $\text{kg}$ , w.w.);  $C_z$  = activity concentration in zooplankton ( $\text{Bq kg}^{-1}$ , w.w.);  $k_{upc}$  = uptake rate of radionuclide to polar cod directly from water column ( $\text{d}^{-1}$ ),  $C_w$  = activity concentration in water ( $\text{Bq l}^{-1}$ );  $C_{pc}$  = activity concentration in polar cod ( $\text{Bq kg}^{-1}$ , w.w.);  $k_{epc}$  = excretion rate from polar cod ( $\text{d}^{-1}$ )

## Trophic level 4: Harp Seal (uptake via food only):

We assume that the uptake of radionuclides directly from the water column to the harp seal is negligible and that the harp seals diet, in simplified terms, consists of 50 % polar cod and 50 % zooplankton.

$$\frac{dC_{hs}}{dt} = 0.5 \cdot (AE_{hs} \cdot IR_{hs} \cdot C_z) + 0.5 \cdot (AE_{hs} \cdot IR_{hs} \cdot C_{pc}) - C_{hs} \cdot k_{ehs} \quad (4.4)$$

$AE_{pc}$  = assimilation efficiency (dimensionless) for harp seal;  $IR_{hs}$  = ingestion rate per unit mass of harp seal ( $\text{kg d}^{-1}$  per  $\text{kg}$ , w.w.);  $C_z$  = activity concentration in zooplankton ( $\text{Bq kg}^{-1}$ , w.w.);  $C_{pc}$  = activity concentration in polar cod ( $\text{Bq kg}^{-1}$ , w.w.);  $C_{hs}$  = activity concentration in harp seal ( $\text{Bq kg}^{-1}$ , w.w.);  $k_{ehs}$  = excretion rate from harp seal ( $\text{d}^{-1}$ )

### 4.1.2 Parametrisation of model

#### 4.1.2.1 Bioconcentration factors for phytoplankton

IAEA's Technical Report 247 (IAEA, 1985) derives a wet weight Cs CF of 20 based on the discussion made by Styron *et al.* (1976). It is interesting in the context of EPIC to note that a wide range of concentration factors were reported by Styron *et al.* (1976) for marine phytoplankton (CF 1 to 403 based on a dry weight basis) in response to changes in temperature (experimental range =  $4\text{-}40^{\circ}\text{C}$ ) and salinity (experimental range = 3.5-44 ppt). Another key parameter that can influence the CF is phytoplankton population growth.

Assuming that marine phytoplankton contain approximately 96 % water (Styron *et al.*, 1976), more recent experimental data reported by Heldal *et al.* (2001) can be transformed to a w.w. CF for  $^{137}\text{Cs}$ . CFs (Dry weight basis) of up to  $1 \times 10^3$  for growing cells and  $2.5 \times 10^3$  for non-growing cells (temperature of  $12 \pm 1^{\circ}\text{C}$  in both experimental setups) were reported by Heldal *et al.* (2001). This converts to a f.w. CF of up to 40 and 100 for growing and non-growing cells respectively. No significant differences in the uptake of  $^{137}\text{Cs}$  between species were observed. For lack of more detailed information the generic values reported in IAEA's Technical Report 247 (IAEA, 1985) have been used for the radionuclides in question. CFs of 20 and  $1 \times 10^5$  have been reported for Cs and Pu respectively.

#### 4.1.2.2 Feeding parameters (IR) for organisms

Food consumption or ingestion rates (normalised to the wet weight of the organism) have been tabulated by Thomann (1981) for different trophic levels. These values are presented in Table 4.1. Polar cod have been defined as a large fish although in reality they probably intersect trophic levels 3 and 4 as defined by Thomann (1981). Adult polar cod may attain lengths of up to 40 cm and weigh several hundred grams.

**Table 4.1** Assumed feeding parameters (Thomann, 1981).

Trophic level	Assumed range of wt (g(w))	Food consumption (IR) (kg/kg-d)
(2) zooplankton	0.001-1	0.105
(3) small fish	0.005-50	0.017
(4) large fish	5-5000	0.009

Innes *et al.* (1987) have provided the following allometric relationship for adult seals:

$$IR \text{ (kg/day)} = 0.079M^{0.71} \quad (4.5)$$

Where  $I_{\text{seal}}$  = rate of biomass ingestion (kg/d, w.w.);  $M$  = Body mass (kg)

Within the project EPIC we have selected a seal with associated mass of 160 kg. Applying this value to Equation 4.5, we derive a (mass-normalized) IR of 0.018. Notably this value is higher than the IR value presented in Table 4.1 for Trophic level 4, but might be accounted for by the fact that homiotherms need to assimilate greater quantities of food in order to maintain body temperatures.

#### 4.1.2.3 Water uptake, excretion rates and assimilation efficiencies

Parameters defining water uptake rates,  $k_u$  ( $d^{-1}$ ), excretion rates  $k_e$  ( $d^{-1}$ ) and assimilation efficiencies, AE (dimensionless) for zooplankton and fish are presented in Table 3.21. The parameter values for Trophic level (4) , large fish, have been taken to be representative of polar cod.

**Table 4.2** Parameters for  $^{239}\text{Pu}$  and  $^{137}\text{Cs}$  (Thomann, 1981).

Trophic level	$^{239}\text{Pu}$			$^{137}\text{Cs}$		
	$k_u$	$k_e$	AE	$k_u$	$k_e$	AE
(2) zooplankton	18.7	0.05	0.01	0.49	0.03	0.5
(3) small fish	0.3	0.02	0.01	0.07	0.003	0.5
(4) large fish	0.01	0.01	0.01	0.01	0.0018	0.5

$k_u$  = Uptake rate from water column ( $d^{-1}$ )

$k_e$  = Excretion rate from organism ( $d^{-1}$ )

AE = Assimilation efficiency for organism (dimensionless)

For the seal, AEs for both  $^{137}\text{Cs}$  and  $^{239}\text{Pu}$  have been set to the same value as that representative of lower levels in the food-chain. The value of  $k_u$  , the direct uptake from the water column, is assumed to be zero.

An allometric relationship may be used to estimate  $^{137}\text{Cs}$   $k_e$  for seal. The following equation has been applied by the USDoE (USDoE, 2002) based on earlier considerations (Whicker & Shultz, 1982).

$$\lambda_i = \frac{\ln 2}{3.5W^{0.24}} \quad (4.6)$$

where  $\lambda_i$  = biological decay constant ( $d^{-1}$ ),  $W$  = mass of animal (g, w.w.).

This yields a value of **0.0112**  $d^{-1}$  for seal. Assuming that excretion is the only process by which the organism loses contamination then the biological decay constant can be set equal to the excretion rate. Although this value has been used in this preliminary version of the model, it is apparent that using elimination rates derived using allometric relationships lead to a more rapid than expected loss from high level predators such as seal. The data in Table 4.2 suggest that  $k_e$  decreases as trophic level increases although this trend may be offset by the fact that mammals are homeothermic with concomitantly higher metabolic rates (for a stated mass). More work will clearly be required in deriving more robust excretion rate data for this radionuclide.

In a similar fashion, a biological half-life can be derived for Pu based on a simple allometric relationship. This is defined as (USDoe, 2002):

$$\lambda_i = \frac{\ln 2}{0.8W^{0.81}} \quad (4.7)$$

where  $\lambda_i$  = biological decay constant ( $d^{-1}$ ),  $W$  = mass of animal (g, w.w.).

This yields an excretion rate of  $5 \times 10^{-5} d^{-1}$  for a  $160 \times 10^3$  g seal. The application of this allometric relation requires further investigation. The incorporation of Pu is complex and will depend on a number of factors including the age of the mammal. Quite different removal rates are likely to be associated with various biological compartments, e.g. blood, muscle, bone etc.

#### 4.1.3 Implementation of model

The compartmental model described above, including concomitant parameter values defined thereafter, has been incorporated into and solved numerically using the Matlab©-based ecosystem modelling tool “ECOLEGO”. The water concentration has been set to unit concentration (of  $^{137}\text{Cs}$  and  $^{239}\text{Pu}$ ) through the whole simulation. Radioactive decay is accounted for during the simulation.

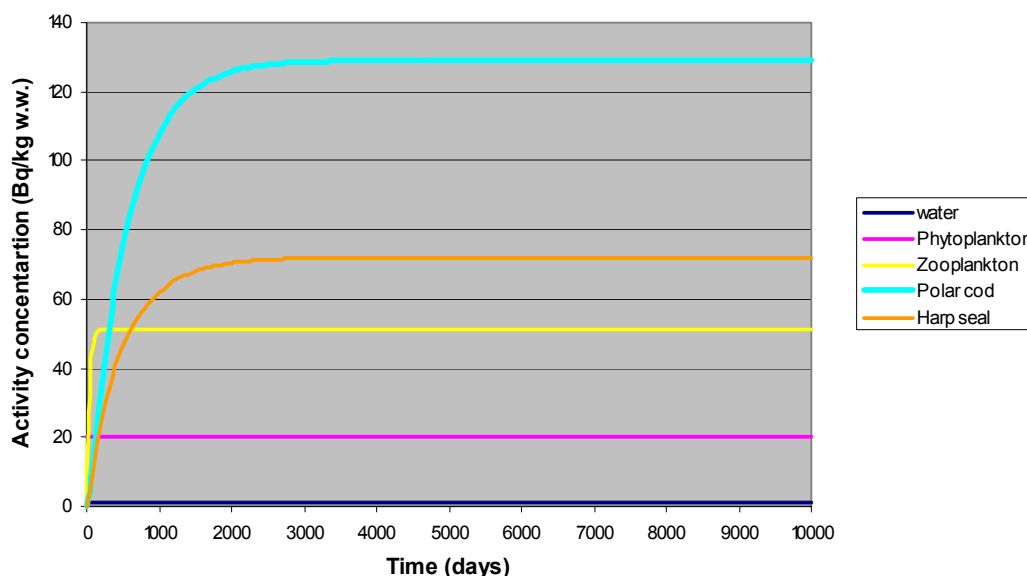
The simulation results using the biokinetic allometric model are shown in Figures 4.2 and 4.3 for  $^{137}\text{Cs}$  and  $^{239}\text{Pu}$  respectively.

For the simulation concerning  $^{137}\text{Cs}$ , the preliminary results suggest that equilibrium is not attained for higher trophic levels, i.e. polar cod and harp seal, before time  $> 2000$  days. This has obvious implications in relation to the interpretation of field data if the activity concentrations in water are changing rapidly with time. Biomagnification<sup>3</sup> appears to occur for the lower trophic levels but is not occurring at the highest trophic level, i.e. seal. It should be noted, however, that the uncertainty associated with the excretion rate ( $k_e$ ) of  $^{137}\text{Cs}$  for seal is large and that this parameter has a significant effect on the equilibrium CF. Setting the  $k_e$  for  $^{137}\text{Cs}$  to  $0.0018 d^{-1}$ , for example, a value attributed to Trophic level 4 (Table 4.2) results in a CF of several hundred for seal. The equilibrium  $^{137}\text{Cs}$  CFs are approximately 50, 130 and 70 l/kg for zooplankton, polar cod and seal respectively. These values appear sensible. They compare to IAEA (Table 3.1) recommended values of 40 l/kg for zooplankton and 100 l/kg for generic fish. The  $^{137}\text{Cs}$  value of 70 l/kg for seal corresponds to a value of 400 l/kg

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<sup>3</sup> Biomagnification defined as an increase in body mass concentration of a contaminant as it passes from low trophic levels to higher ones

included in Table 3.2 for a generic pinniped (a close match considering the very few data upon which the IAEA value is based).



**Figure 4.2** Activity concentrations (w.w.) of  $^{137}\text{Cs}$  for selected marine organisms derived from biokinetic allometric modelling. The simulation was run for a unit activity concentration in water.

Several points of interest arise from the simulation for  $^{239}\text{Pu}$  (Fig. 4.3). Transfer to successively higher trophic levels is low – there is a fall of several orders of magnitude between primary producers, represented by phytoplankton, and polar cod representing trophic level 3-4. It should be noted, however, that the model predicts that this decreasing trend in activity concentrations along the food-chain is reversed for the highest trophic level, represented by seal. The simulated results for seal display activity concentrations in the region of 2 orders of magnitude higher than those observed for polar cod (one of its prey species) once the system has equilibrated. This prediction is strongly influenced by the fact that the other component of the seal’s diet, zooplankton, has a much higher activity concentration associated with it. Equilibrium is attained very slowly for “seal” (reflecting, in part, the very low, allometrically-derived excretion rate). In this case, equilibrium is only truly obtained after  $6 \times 10^4$  days (165 years) of simulation. Clearly, equilibrium, even in the unlikely circumstance where water concentrations remain unchanged over highly protracted time scales, is unlikely to be attained over the life-time (in the order of decades) of the seal. The model predictions compare quite favourably with the recommended values reported by the IAEA. The equilibrium CFs of  $2.5 \times 10^3$  and 25 l/kg predicted from model runs for zooplankton and polar cod compare with IAEA (Table 3.1) recommended values of  $4 \times 10^3$  and 100 l/kg for zooplankton and generic fish respectively. For seal, as discussed above, equilibrium is not reached between the water and seal body compartments over the life time of the organism. A true equilibrium of  $4.5 \times 10^3$  l/kg is attained after 165 years, however, following a 1 year equilibration period, a concentration ratio of approximately 75 l/kg is predicted. This latter value compares with the empirically-derived value presented in Table 3.2 of 8 l/kg for pinniped liver. The appropriateness of applying a Pu CF value to a high level predator, like seal, is clearly open to question.



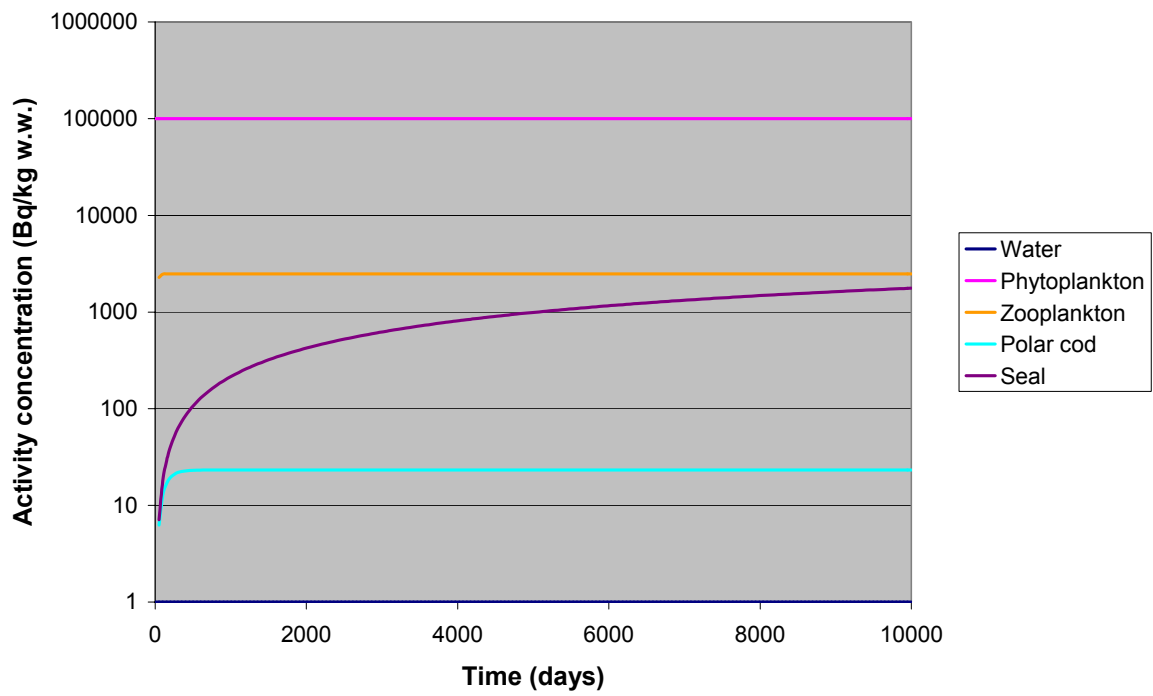


Figure 4.3 Activity concentrations (w.w.) of  $^{239}\text{Pu}$  for selected marine organisms derived from biokinetic allometric modelling. The simulation was run for a unit activity concentration in water.

## 5. Conclusions

A consideration of the types of contaminant transport model applied by institutes across Europe indicates a wide variability in type and applicability. Key model types, defined by structure include time differential, OD or box models and time and space differential, 1D, 2D and 3 D models. Some basic elements are common to most of the available models including the ability to simulate hydrodynamic processes, contaminant advection and dispersion and, in a more limited number of cases, to simulate sediment processes including the transport of sediment-bound contamination. Arctic-specific model are, understandably, less ubiquitous. In the context of Arctic marine impact assessments, a number of processes that may be unimportant for temperate sea areas, may require consideration in this special case. For example, it may be necessary to model contaminant interactions with and transport by ice. In view of the coordinating institute's experience with simulating the behaviour of radionuclides in Arctic environments, the NRPA's box model has been presented to demonstrate the applicability of transport modeling in an assessment context. The model simulates radionuclide-marine processes including advection between boxes, interaction with sediments, pore water diffusion, bioturbation, sediment mixing and burial. The output of data from this model in the form of contaminant levels in water, suspended and deposited sediments can be used to derive external dose rate levels through the application of relevant dosimetric models. The model also links to the biological component of the ecosystem through the use of concentration factors allowing the simple mapping of an organism's body concentration to the ambient sea water concentration. In the simulation example given, Arctic specific CFs, derived from work in the EPIC project, have been used.

Biological transfer is often modeled using the CF approach. A large dataset is available for use providing information for generic organism groups from temperate sea areas, through the IAEA (IAEA, 1985 and updated version of this document). However, the applicability of these data to environmental impact assessment generally are hindered from the facts that (i) the IAEA report was collated for human impact assessments and is therefore biased towards edible organisms and the edible parts of those organisms and (ii) the report contains little information on the distribution of contaminants within species, details for which may be necessary in the fulfillment of robust dose calculation. Data have, therefore, been collated from the open literature and are recommended to be used as supplementary information to the data present within the aforementioned IAEA report (see Table 3.6-3.19).

In further consideration of the CF approach, 2 major problems become evident. The first problem relates to the fact that, under normal field conditions, the abiotic and biological compartments of the environment may not be under equilibrium. Activity concentrations in water can change rapidly in response to variable contaminant discharge regimes and relatively short water "flushing" times. Biological uptake and depuration rates are normally low, i.e. half-times are large, when compared with these physical process rates. CFs may therefore poorly characterize the water-biota partitioning. The second problem arises simply from the fact that CF data are not available for all radionuclide-biota combinations. In both cases dynamic model might allow more robust prognoses to be made in the course of an impact assessment. A biokinetic model has been developed at the NRPA specifically for an Arctic pelagic marine food-chain and with "reference" organism groups in mind. In this initial study the model has been applied with the objective of deriving equilibrium CFs for sea mammals – a organism group for which few empirical data exist. Allometric relationships have been used in several cases where empirical data were unavailable for parameterization. The preliminary model appears to give reasonable predictions for  $^{137}\text{Cs}$  and  $^{239}\text{Pu}$  and demonstrates the fact that high trophic level organisms may take very long time periods to become equilibrated with ambient water concentrations.

It would be clearly advantageous to link the available marine transport model, i.e. the NRPA box model (Chapter 2) to the dynamic model discussed in Chapter 4. Such an operation is possible, but further work is required in defining the assumptions that are necessary for this to be practicable and for defining

under which circumstances the application of the model would be valid. In other words, coupling the models would be premature before further model testing is undertaken.

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**StrålevernRapport 2003:1**  
Virksomhetsplan for 2003

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Utslipp av radioaktive stoffer fra Sellafield-anleggene  
En gjennomgang av britiske myndigheters  
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**StrålevernRapport 2003:3**  
MOX, En del av kjernebrenselsyklusen

**StrålevernRapport 2003:4**  
LORAKON  
Resultater fra Ringtest i 2000 og 2001

**StrålevernRapport 2003:5**  
Monitoring of  $^{99}\text{Tc}$  in the Norwegian Arctic marine environment

**StrålevernRapport 2003:6**  
Treårig tilstandsrapport for konsesjonsbelagte anlegg ved  
Institutt for energiteknikk