Radionuclides in the Baltic Sea
Ecosystem models and experiments on transport and fate

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Abstract

Manmade radionuclides have been introduced to the environment for almost a century. The main source has been the nuclear weapons testing programmes, but accidental releases from the nuclear power production industries have also contributed. The risk to humans from potential releases from nuclear facilities is evaluated in safety assessments. Essential components of these assessments are exposure models, which estimate the transport of radionuclides in the environment, the uptake in biota, and transfer to humans. Recently, there has been a growing concern for radiological protection of the whole environment, not only humans, and a first attempt has been to employ model approaches based on stylised environments and transfer functions to biota based exclusively on bioconcentration factors. They are generally of a non-mechanistic nature and involve no knowledge of the actual processes involved, which is a severe limitation when assessing real ecosystems.

The research presented in this thesis attempts to introduce a methodology for modelling exposure of biota that is based on systems ecological theories and concepts. All presented papers concern bioaccumulation and circulation of radionuclides in coastal areas of the Baltic Sea, which is a sea surrounded by several nuclear power plants, waste repositories and reprocessing facilities. Paper I illustrates how an ecosystem model can be used to predict the fate of C-14 in a bay, and to explore the influence of uptake route and water exchange on the concentrations in biota. Due to the longevity of many radionuclides, time spans of thousands of years need to be considered in assessments of nuclear waste facilities. In Paper II, the methodological problems associated with these long timescales are discussed and a new modelling approach is proposed. An extension and generalisation of the C-14 flow model into a generic model for other radionuclides is described and tested in Paper III. This paper also explores the importance of three radionuclide specific mechanisms (plant uptake, excretion and adsorption to organic surfaces) for the concentrations in biota. In Paper IV, the bioaccumulation kinetics of three radionuclides in three key benthic species of the Baltic Sea is studied experimentally. Paper V considers remobilisation and redistribution of sediment-associated radionuclides due to biological mixing, in a microcosm study.

The findings in this thesis show both that it was possible to use an ecosystem approach to assess the exposure to biota, and that this approach can handle many of the problems identified in the use of traditional exposure models for radionuclides. To conclude, frameworks for the protection of the environment from ionising radiation would benefit from implementing methodologies based on ecologically sound principles and modelling techniques.
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List of Papers

This thesis is based on the following Papers, which will be referred to in the text by their Roman numerals:


III. **Kumblad L**, Kautsky U and Næslund B. Transport and fate of radionuclides in aquatic environments - the use of ecosystem modelling for exposure assessments of nuclear facilities. (*Manuscript*).

IV. **Kumblad L**, Bradshaw C and Gilek M. Bioaccumulation of Cr-51, Ni-63 and C-14 in Baltic Sea benthos. (*Submitted to Environmental Pollution*).

V. Bradshaw C, **Kumblad L**, and Larsson A. Remobilisation of buried radionuclides from anoxic sediments by bioturbation. (*Manuscript*).

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Introduction

Radioactivity is a universal phenomenon that has existed since the beginning of time, but was first described in 1896 when Henri Becquerel confirmed that uranium salts generated ionising radiation. The era of artificial radioactivity started soon thereafter as radioactive isotopes were artificially produced. In 1942, the world truly entered the atomic age when a team of scientists for the first time initiated and controlled a self-sustaining nuclear chain reaction. These experiments paved the way for the large-scale production of anthropogenic radionuclides in nuclear reactors and the development of nuclear weapons. Only three years later, a nuclear test explosion in New Mexico caused the first wide-ranging radioactive contamination of the environment (Aarskog 1994). Since then, man-made radioactivity has entered the environment from a variety of activities (MacKenzie 2000). At present, there are thousands of sites around the world that are measurably contaminated with artificial radionuclides (Whicker et al. 1999).

Ionising radiation has the ability to penetrate living tissues and therefore has the potential to cause severe damage to biota and ecosystems in two ways: (i) internal radiation from internally stored radionuclides received from the environment via ingestion or diffusion, and (ii) external radiation from radionuclides in the surroundings. The injuries of significance caused by ionising radiation concern the destruction of DNA, which may result in mutations, cancer or cell death. Mutations in germ cells are particularly harmful as they may be inherited by descendents and cause effects at the level of population or ecosystem (Harrison and Knezovich 2001). Due to the long half-life of many radionuclides even low emissions pose a risk. This is because environmental processes such as sedimentation and bioaccumulation may increase the concentration of contaminants in certain parts of the environment to such extent that they may be harmful for living organisms (Kautsky and Hedin 1998).

Radioecology is the study of the transport, fate, and effects of radionuclides in the biosphere. It was first introduced as a separate field of science by Eugene Odum in 1956. Since then, radioecological research has considerably increased the knowledge of radionuclide behaviour in the environment. It has also widened the understanding of ecological processes through the use of
radioactive isotopes as biological tracers (Whicker and Pinder 2002). Many modelling tools used in the assessment of radionuclide transport and fate in ecosystems are still, however, very simplistic and non-dynamic in nature (e.g. Thiessen et al. 1999).

The Papers included in this thesis introduce systems ecological theories and methodologies into exposure modelling of radionuclides. This methodology suggests that a holistic approach is needed to accurately describe the behaviour of radionuclides in complex and highly dynamic systems, especially when long time-periods need to be considered. All Papers concern bioaccumulation and circulation of radionuclides in coastal areas of the brackish Baltic Sea, which is a semi-enclosed sea surrounded by several nuclear power plants, waste repositories and reprocessing facilities (HELCOM 1995; Nielsen et al. 1999; HELCOM 2003) (Figure 1). The Baltic Sea has also been polluted with radionuclides from the Chernobyl accident, nuclear weapons test, nuclear waste dumping and nuclear facilities (HELCOM 1995; Nielsen et al. 1999; Elmgren 2001; Wallberg and Moberg 2002; HELCOM 2003).

**Paper I** illustrates how an ecosystem model can be used to predict the fate of C-14. The constructed model was also used to explore how changes in the uptake pathway and in the rate of water exchange would influence the C-14 concentration in biota.

In **Paper II**, predicted future changes in environmental parameters were tested by re-scaling the ecosystem model (Paper I). This was performed to evaluate the reliability of the ecosystem modelling approach for assessing future potential radionuclide releases from, for example, nuclear waste repositories.

**Paper III** focused on the extension and generalisation of the C-14 flow model (Paper I) into a generic radionuclide model for other radionuclides. Paper III also explores the importance of three radionuclide-specific mechanisms (plant uptake, excretion and radionuclide adsorption to organic surfaces) for the concentrations in biota and other components of the system.

In **Paper IV**, the bioaccumulation kinetics of three radionuclides in three key benthic species of the Baltic Sea was studied experimentally.

**Paper V** considers remobilisation and redistribution of sediment-associated radionuclides due to biological activity, bioturbation, in a microcosm study.
Figure 1. The Baltic Sea drainage basin and location of nuclear reactors and dumping sites for radioactive waste in the area. Also shown are the locations of Öregrundsgrepen, where the modelling studies (Papers I, II and III) were performed, and the Askö field station, where the experimental studies (Papers IV and V) were carried out.

**Ecological Risk Assessments & Safety Assessments**

The likelihood that adverse effects on humans and the environment will occur, are occurring, or have occurred from discharges of contaminants is evaluated in ecological risk assessments (Suter 1995). Those assessments have also become an important tool in decision-making processes for managing environmental problems (Suter 1993; US-EPA 1998). To support decisions about the localisation and performance of nuclear facilities, safety assessments are performed (Chapman and McCombie 2003). These mainly aim to evaluate the safety of humans in relation to the performance of the facility, but have
occasionally also included ecological risk assessments to evaluate potential
doses and effect on biota (e.g. Goodwin et al. 1996).

Usually, ecological risk assessments are composed of several associated
components (Table 1). First, there is a hazard definition where the environment
is described, the assessment endpoints are identified and the source terms
estimated. Then an exposure assessment characterises the properties of the
contaminant, determines the transport and fate of the contaminant in the
environment and estimates the degree of uptake and external exposure. In the
effect assessments, the relationship between the degree of exposure and the
nature, severity and duration of the effects is established. Finally, in the risk
characterisations, the exposure and effect assessments are combined and an
evaluation, explanation and conceptual context for the results provided (Suter
1995). Important issues in these assessments are also to identify the critical
variables in the assessment, and estimate their major uncertainties. The
procedure to attain predictions and estimations is highly dependent on
modelling and often involves a number of different model types to meet the
aims of the different components of the risk assessment. The Papers included
in this thesis consider several of the stages of an ecological risk assessment:
description of the ecosystem, estimation of concentrations in abiotic and biotic
components and doses to biota (Table 1).

Table 1. Components included in ecological risk assessments of environmental impacts of
contaminants. The assessment products from an assessment of radionuclides (RN) released to
an aquatic ecosystem are chosen as an example.

<table>
<thead>
<tr>
<th>Assessment components</th>
<th>Stage of assessment</th>
<th>Product in an assessment of radionuclides (RN)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Hazard definition</td>
<td>1. Description of the ecosystem</td>
<td></td>
</tr>
<tr>
<td></td>
<td>2. Identification of endpoints</td>
<td></td>
</tr>
<tr>
<td></td>
<td>3. Source term characterisation</td>
<td>RN-conc. in water</td>
</tr>
<tr>
<td>Exposure assessment</td>
<td>4. Distribution modelling</td>
<td>RN-conc. in abiotic components</td>
</tr>
<tr>
<td></td>
<td>5. Exposure modelling</td>
<td>RN-conc. in biota</td>
</tr>
<tr>
<td></td>
<td>6. Dose calculations</td>
<td>Dose to biota</td>
</tr>
<tr>
<td>Effect assessment</td>
<td>7. Dose effect relationships</td>
<td>Effects on individuals</td>
</tr>
<tr>
<td>Risk characterisations</td>
<td>8. Environmental effect</td>
<td>Effects on populations and ecosystem structure and function</td>
</tr>
<tr>
<td></td>
<td>assessment: evaluation and extrapolation of exposure and effect assessment</td>
<td></td>
</tr>
</tbody>
</table>
Despite the important role that risk/safety assessment models often play in supporting decisions about contamination issues, many rely on very simplistic approaches, and fail to incorporate basic ecological information and modelling capabilities (Pastorok 2002). This is the case for assessments of radionuclides. Until recently, the focus has exclusively been the protection of humans (e.g. Strand and Larsson 2001; Copplestone et al. 2004) and thus the primary concern has been the pathways leading to human consumption in accordance with the belief that “if man is adequately protected, then other living organisms are likely sufficiently protected as well” (ICRP 1977; ICRP 1991). As a result, ecological components have often been omitted or incompletely described (e.g. IAEA 1999a; Vieno and Nordman 1999). The limitation to human health protection is now increasingly questioned, and new regulations require that effects on ecosystems should be considered (e.g. SSI 1998; IAEA 1999b). These requirements have, among others, initiated the development of new assessment frameworks (e.g. EPIC 2001; Strand and Larsson 2001) and the establishment of a new task group focused on the protection of the environment from ionising radiation by the International Commission on Radiological Protection, ICRP (Holm 2004).

*Exposure assessment of radionuclides*

Distribution and exposure models are essential components of the assessments of any contaminant. Their role is to determine the main transfer pathways for the contaminant in the ecosystem, identify organism groups that may become highly exposed, estimate uptake and elimination dynamics, retention times in biota and the ecosystem (Suter 1993). The structure of exposure models for radionuclides varies, but most are of a non-mechanistic nature and developed for steady-state conditions (Thiessen et al. 1999; Whicker et al. 1999). The environmental distribution modelling has mostly been performed using transfer factors such as bioconcentration factors (BCF) and distribution coefficients (K_d) (Ribbe et al. 1991; Nielsen et al. 1995; Osvath et al. 1999; Toscano-Jimenez and Garcia-Tenorio 2004). These factors are empirically derived ratios of radionuclide concentration in organisms or in/on particles to the concentration of dissolved radionuclides in the water.
BCF-based models are easy to use but are open to criticism for various reasons:

- BCFs do not involve any fundamental understanding of uptake and transport processes in the environment, as they are empirically derived from laboratory studies or field measurements (Sansone et al. 2002).
- BCFs are only valid if steady-state conditions in the system can be assumed, which is not always the case (Whicker et al. 1999; Thorne 2003).
- The relationship between the radionuclide concentrations in the environment and the radionuclide concentration within an organism may not be linear (Brown et al. 2003)
- BCFs are not available for all organisms and specific environments of interest in assessments for nuclear facilities (Jones et al. 2003b).

The first three Papers in this thesis explore the possibility of using an ecological modelling approach to overcome some of the problems related to the use of BCF-based models.

**Ecological Modelling**

Ecological models describe or predict ecological processes or endpoints beyond the individual organism level; e.g. population abundance, community species richness, productivity/distributions of organisms, or the transfer of energy (e.g. Odum and Hoskin 1958; Odum 1983). Usually one distinguishes between three different types of ecological models: (i) population models, (ii) ecosystem models and (iii) landscape models (Fitz et al. 1996). Population models typically deal with the dynamics and abundance or distribution of single species, whereas ecosystem models describe ecological systems composed of interacting species. Ecosystem models typically include food web modelling, are often of mass balance type and consider both abiotic and biotic components of the system. Because of the inherent complexity of such models, they tend to be specific either to a geographic area or to a type of ecosystem. The third class of ecological model is the landscape model that either is a spatially explicit ecosystem model (e.g. Fitz et al. 1996), or considers how populations are occupying the landscape (e.g. Hanski 1999).
One purpose of using ecological models in risk assessments may be to extrapolate a measurement endpoint to an assessment endpoint (Pastorok and Akcakaya 2002). The assessment endpoints are the environmental characteristics or values that are to be protected, e.g. species diversity or ecosystem productivity, and the measurement endpoints are quantitative expressions of observed or measured biological responses, such as mortality or fecundity. Ecological models may also be used to get an overview of complex systems and to understand ecosystem behaviour (Jørgensen 1995), or for fate modelling of nutrients, energy and pollutants in the environment (e.g. Newell et al. 1982; Jansson and Jansson 1988; Jackson 1996; Gilek et al. 1997; Morrison et al. 1997; Bartell et al. 1999; Heymans and Baird 2000; Naito et al. 2002). Due to the mechanistic nature of most ecosystem models it is possible to explore how the ecosystem responds to changes in environmental variables (Bartell et al. 1999; Paper II). This provides a method to evaluate the environmental fate of future contaminant releases (tested and discussed in Paper II).

Mass balanced ecosystem models of aquatic food webs provide a way to analyse how resources are linked to consumers through fluxes of e.g. carbon and nutrients (Kremer and Nixon, 1978). For instance, in the simplest case of a mass balance model for carbon in an aquatic ecosystem, the model consists of one compartment with a total inflow of carbon to the system that is balanced by export, total respiration and total burial of carbon in sediments. This approach was used by e.g. Wulff et al (2001) when describing nutrient circulation in the Baltic Sea. This type of modelling can also be applied for single organisms or trophic interactions between functional groups (e.g. Sandberg et al. 2000; Paper I, II and III). Mass balance modelling at this level often assumes that consumption should equal the sum of production, respiration, and egestion (unassimilated food/faeces) (Crisp 1971). In cases when the estimated inflow of matter exceeds the estimated outflow, there is an indication of biomass increase in the population, e.g. growth or reproduction. In the models presented in this thesis, the modelled outflow variables were production (biomass available for grazing or predation), respiration, faeces, and excess production (growth, death, reproduction) (Figure 2).
Figure 2. Main metabolic processes of heterotrophic organism groups modelled in this thesis. Consumption equals the sum of production, respiration, faeces and excess.

*Using ecosystem models in safety assessments*

The uptake and transfer of radionuclides in the food chain is influenced by a wide range of environmental factors that vary substantially between different ecosystems. As a result, it may seem impossible to handle the great complexity that mechanistic fate models have to consider. It is, however, important to note that the mechanisms by which a radioactive element is incorporated into biological systems are basically the same as those by which the organisms obtain their nutrients from the atmosphere, soil, water or food (Alexakhin et al. 2001). Mineral and nutrient cycles may therefore provide the first important clue of how radionuclides behave in the environment (Whicker and Schultz 1982). Radioactive isotopes generally show similar environmental behaviour as their stable counterparts, although the potential impacts on organisms are enhanced by their ability to emit radiation (Szefer 2002). The chemical behaviour of most radionuclides is also similar to other elements in the same groups of the periodic table (e.g. Klement 1982; Whicker and Schultz 1982). Known properties for stable analogues and/or similar elements may thus be utilised for modelling purposes. For instance, Sr-90 and Ra-226 have similar properties and biogeochemical pathways in the environment as Ca, and Cs-isotopes as K, and in some metabolic processes as Ca (Shapiro et al. 1993). Similarly, the behaviour of tritiated water can be predicted accurately from the hydrological cycle and C-14 from the carbon cycle (Murphy Jr 1993; Paper I). Although the use of stable analogues might provide a concept for modelling of many radionuclides, there are still many radionuclides that do not have any stable counterparts for which the environmental behaviour has been studied. For these, the intrinsic properties of the receiving environments may be used to evaluate their fate, since energy flows determine the rates at which all radionuclides will eventually be partitioned in the environment (Whicker and Schultz 1982). The circulation of energy within an ecosystem, consumption
rates and accumulation constraints by organisms can thus provide a method to estimate possible ranges for radionuclide accumulation. This concept was used in the three modelling studies of this thesis (Papers I, II, and III).

**Exposure models developed in this thesis**

The overall purposes of the modelling studies were (i) to examine the possibility of applying an ecosystem approach to predict the transport and fate of radionuclides released to the environment, and (ii) to evaluate if this approach provides a way to overcome the problems associated with the use of steady state models based on BCFs (as discussed earlier). The models are site specific and focused on radionuclides but can, with some adjustments and new site-specific data, be applied for other contaminants and for other aquatic environments with similar properties.

The models were primarily developed for a safety assessment project of the final repository for radioactive operational waste (SFR) located in the bedrock under the seabed in Öregrundsgrepen, Baltic Sea. The aim was to predict the fate of hypothetically released radionuclides from the repository into the bay above (Figure 3). The development of the models was also related to an international project aiming for a general framework for ecological risk assessments of radionuclides (FASSET).

*Carbon flow model*

The base for any ecosystem model is a detailed description of the environment including both abiotic and biotic components. For the models presented in Papers I and II (for C-14), and III (for 25 radionuclides) the base was a carbon budget compiled from field measurements in the area, that was combined into a carbon flow model for this particular site (described in detail in Paper I). The carbon flow model describes biomasses and carbon dynamics of the ecosystem. It was conceptualised according to Figure 4, although the carbon budget was compiled at a more detailed level.
Figure 3. Map: Location of the study area and the SFR-repository, Öregrundsgrepen (Baltic Sea). Photo: The area to which the models in this thesis project have been applied. The Forsmark nuclear power plant is in the background and the SFR-repository in the foreground.
The main processes in the model were primary production, respiration, consumption and egestion (unassimilated food). The initial data for these parameters were determined at species or family level, but combined to functional groups before they were used in the model. Seasonal variations were taken into account by including temperature and light intensity variability over the year. The carbon flows were constrained by temperature, light intensity and inorganic carbon available for photosynthesis. The biomass was assumed to remain constant between years.

**Figure 4.** Structure of the food web used in the carbon flow model, including the abiotic compartments DIM (dissolved inorganic matter) and POM (particulate organic matter). The arrows in the figure illustrate the flow of organic matter in the ecosystem as well as the flow of hypothetical discharges of radionuclides from the final repository for radioactive operational waste (SFR). The thickness of the lines indicates the relative magnitude of the carbon flows.
To model import and export of matter to and from the system, a water exchange mechanism was connected to all pelagic components, i.e. phyto- and zooplankton, dissolved inorganic matter (DIM) and particulate organic matter (POM). Inorganic carbon was assumed to enter the food web through photosynthesis (primary production) and be transferred to higher trophic levels via grazing and predation. The carbon inflow to the compartments, i.e. primary production or consumption, was balanced by the carbon outflow, i.e. respiration, grazing or predation, and egestion. Egested material and ungrazed primary production was connected to the POM compartment, which provided detritus-feeding organisms (soft bottom fauna) with organic carbon. Carbon lost in respiration was linked to the DIM compartment that served as a recirculation pathway for carbon within the model.

Analysis of the carbon dynamics of the area demonstrated self-sufficiency of carbon resulting in a net export of organic carbon corresponding to almost 10% of the annual primary production. The most significant flow was the export of biomass produced in the phytobenthic and pelagic communities down to the soft bottom and away from the area through water movement.

The carbon flow model was validated in two steps: first against carbon flow models constructed for the area around the island of Askö in the Trosa Archipelago, Baltic Sea (Paper I), and then by incorporating nutrient dynamics (Paper II). The outcomes from these two validations show that the model provides a good representation of the carbon flow in the modelled ecosystem, although some deviations were found for benthic macrofauna. Compared to the Askö area, the biomass of the benthic macrofauna was approximately 5 times lower in this budget. The C:N:P mass balance analyses indicated that the rate of primary production was overestimated and that the food selection for fish could be redistributed.

*C-14 flow model*

C-14 is a radionuclide of considerable interest in disposal of nuclear low-level waste because of the large quantities often found, its high bioavailability, ecological relevance, environmental mobility and relatively long half-life (Liepins and Thomas 1988). In Paper I, the carbon flow model was adapted and applied to modelling of hypothetically released C-14 from the SFR-repository.
In the model, the C-14 isotope was assumed to have the same chemical properties as stable carbon (although a fractionation from air to biota about 0.9 is known (NCRP 1985; Kennedy and Krouse 1989), and to enter the ecosystem through the sediment in a bioavailable form, i.e. either as $^{14}$CO$_2$, H$^{14}$CO$_3^-$ or $^{14}$CO$_3^{2-}$ or transformed from CH$_4$ by microbes. C-14 was released into the model ecosystem at a constant rate during a 1000-year period, introduced into the food web via photosynthesising organisms and then channelled through the system in proportion to stable carbon.

Three types of model analyses were performed: effect of water exchange rate, influence of entry route into the food web, and overall distribution in the environment. The water exchange rate was found to be of great importance for the exposure of the organisms. The greater the water exchange, the quicker C-14 was removed from the area, thus decreasing the concentrations in the compartments. The extent to which the water exchange influenced the C-14 concentrations in the compartments mirrored their location in the ecosystem, their trophic level and how their food sources were affected. The water exchange rate also influenced the time needed for the system to reach steady-state. The entry route for C-14 was shown to be of great importance for the exposure of biota. The concentrations were up to 800 times higher in simulations where the C-14 entered the benthic primary producers directly, compared to simulations where the isotope was equally available for all primary producers. Since the modelled ecosystem is characterised by a high water exchange rate, more than 99% of the discharged C-14 was immediately exported from the system and 0.02% was lost at the air-sea interface. The percentage accumulated by plants and transferred to higher trophic levels was 0.2%, of which approximately 5% was re-circulated in the system via respiration and 74% was retrieved in excess by biota. Almost 10% of the excess was consumed by benthos and 0.25% was buried in the sediment. The rest was exported from the area by water movement.

The results in Paper I demonstrate that it was possible to analyse and numerically describe the transport and fate of discharged C-14 in a whole ecosystem. The method also facilitated an evaluation of how changes in environmental and ecological factors might affect the behaviour of C-14 released to the ecosystem. It may thus be concluded that the ecosystem modelling approach overcomes many of the problems identified in BCF-based
Assessing long-term fate of radionuclides

Assessments of nuclear waste facilities need to consider time spans of thousands of years. This is due to the long half-life of many radionuclides and because the risk of potential releases from repositories may well increase in the distant future (van Dorp et al. 1998). Potential releases are also expected to occur at low level over long periods of time (Klos et al. 1998). This time perspective is far longer than in most other assessments. Paper II discusses the methodological problems associated with this and proposes a method to estimate the fate of potential future radionuclide discharges to the aquatic environment.

A potential release of radionuclides can occur within the next 10,000 years from the SFR-repository (Lindgren et al. 2001). During this period, the shoreline displacement due to land-rise and sea level rise was identified as the most important process for the ecosystem development of this particular area (Brydsten 1999). This process will transform the coastal ecosystem to freshwater and then to terrestrial environments (Figure 5).

Figure 5. Study area above the final repository of radioactive operational waste (SFR) in Öregrundsgrepen (Baltic Sea) today and 2000 years from now showing the type of ground cover (modified from Brydsten 1999).

The mechanistic nature of ecosystem models facilitates the study of the individual processes involved and thus allows modification and scaling of...
model parameters and functions due to predicted changes in environmental conditions. The modelling methodology described in Paper I can increase the possibility of assessing risks of radionuclide releases in the far future. This was tested in Paper II.

The method used to predict the fate of potential future discharges of C-14 to the bay above the SFR-repository was to normalise and scale the distribution and flows of organic matter in the ecosystem at the site today for predicted changes in bathymetry (Brydsten 1999) and water exchange regimes (Engqvist and Andrejev 2000). The ecosystem model for the area today (Paper I) was used as a platform for the extrapolation to a separate carbon flow model for the ecosystem 2000 years from now (Paper II). Assumptions required for the scaling included (i) that the ecosystem will retain the same functional organism groups, and (ii) that these groups will remain in the same habitat (depth intervals) and have the same densities within the habitat. It was also assumed (iii) that the light intensity and temperature will remain constant on an annual basis and (iv) that the water transparency will not decrease more than 50%.

Comparisons of the carbon dynamics in Papers I and II indicates that the phytobenthic community will clearly dominate the area also in the future (90% of the biomass), which is a consequence of the shallower water in the area (Figure 6). The biomass distribution is expected to change from equal occurrence of plants and animals (biomass basis) to an environment dominated by plants (86%).

The scaled ecosystem model was used to make a long-term prediction of the consequences of a release of C-14 2000 years from now. The changed ecosystem structure affected the carbon fluxes, and thus the C-14 fate in the ecosystem. A release of C-14 to the modelled ecosystem 2000 years from now would cause almost a thousand-fold increase in biota concentrations compared to a hypothetical release today.

Paper II showed that it was possible to re-scale an ecosystem model into a new model that describes the expected properties of a future ecosystem in a realistic way. The scaling includes many uncertainties but still provides a way to both qualitatively and quantitatively assess the likely effects of discharges of contaminants in the future.
Figure 6. The amount ($10^6$ g C) and proportion (%) of total biomass between communities in the study area today and 2000 years from now as modelled in Papers I and II. The phytobenthic community comprises benthic plants, macrograzers, filter feeders, benthic micro- and meiofauna, the soft bottom community filter feeders, benthic macro-, micro- and meiofauna, and the pelagic community phytoplankton, zooplankton and fish.

The use of ecosystem models to assess the fate of other radionuclides

In safety assessments of nuclear facilities, not only C-14 but a multitude of different isotopes need to be considered (Jones et al. 2003a). Paper III attempts to extend and generalise the C-14 model (Paper I) into a generic radionuclide model for the ecosystem above the SFR-repository. The objective was, again, to test the feasibility of the ecosystem modelling approach for exposure modelling of radionuclides having different chemical and physical properties, and to explore how different mechanisms influence the radionuclide fate.

In the model presented in Paper III, the radionuclides were, as for C-14, assumed to follow the flow of organic matter in the system but also regulated by radionuclide-specific mechanisms. These mechanisms were: (i) active radionuclide uptake by plants, (ii) excretion of radionuclides by animals, and (iii) adsorption of radionuclides to organic surfaces.

The performance of the model was tested by calculating BCFs from the modelling results for 25 different radionuclides and comparing these with
empirical BCFs published by the International Atomic Energy Agency (IAEA 1985). In general, the BCFs calculated in Paper III were in the same order of magnitude as the IAEA BCFs for most organism groups and radionuclides. This suggests that the model produces comparable results to empirical radionuclide data, even though the only radionuclide specific input parameters used in the simulations were adsorption efficiencies ($K_d$) and bioconcentration factors for plants (BCFs). The remaining transfer processes were exclusively driven by the carbon flow model.

Analyses of how the water exchange rate affected the radionuclide fate in the system showed that the water exchange rate greatly affected the exposure to biota, both by dilution of the radionuclide concentration in the water and by diluting plankton organisms. This was also concluded in Paper I. The influence of the three radionuclide specific mechanisms on the radionuclide fate was also explored. Analyses of the excretion rate indicate that the biomagnification potential of radionuclides in all organism groups was not larger than a factor 4. When comparing results from simulations with and without surface adsorption of radionuclides to the animals, the surface adsorption mechanism was found to be more important than the plant uptake mechanism for most organisms, although they were equally important for the exposure of fish. Most of the internal body content of radionuclides originated from ingestion of surface-associated radionuclides. This emphasises the importance of food ingestion for the radionuclide exposure, even for radionuclides having low BCF values. These results also demonstrate that the BCF concept, i.e. radionuclide uptake directly from water, is misleading, since food requirements and trophic interactions of organisms seem to be of considerable importance to be able to accurately determine the exposure.

Uncertainties associated with the estimated ecosystem data was evaluated in probabilistic analyses. Probabilistic analyses are a method often used in safety assessment of radionuclides (Klos et al. 1998; Karlsson et al. 2001; Zeevaert et al. 2001; Higley et al. 2003), but have only been used occasionally for ecosystem models (e.g. Bartell et al. 1983). In a probabilistic analysis it is possible to assign distributions and a variation range to selected model parameters and to obtain statistics from simulations where the parameter estimations were randomly sampled from the assigned distributions. Thirty ecological input parameters were selected for the probabilistic simulations in
Paper III. The analyses showed that the majority of the modelling results varied less than one order of magnitude within the 5 and 95 percentiles. This suggests that a well structured ecosystem model with realistic parameter estimations will produce reliable results even if some ecological input parameters, such as the biomass or the metabolic rate of an organism group, are uncertain.

A sensitivity analysis of the 30 selected ecosystem input parameters was also performed. This analysis showed clearly that the exposure of an organism was primarily affected by the properties of the organism itself, such as consumption rate and size. However, the sensitivity analysis did not include the effect of water turnover and the influence of the uptake and elimination constants, which all have great importance.

The use of ecosystem models to assess dose to biota and humans

In the context of ecological risk assessments of radionuclides, the objective of the exposure assessment is to facilitate dose estimations to biota. The absorbed dose is the mean energy imparted by ionising radiation to the tissue per unit biomass (Gy = J kg wet weight\(^{-1}\)) (Cooper et al. 2003). At present, there is no internationally accepted methodology for dose calculations to biota (e.g. Strand and Larsson 2001; Chapman and McCombie 2003), and few safety assessments for radionuclides have included dose calculations to biota. The models presented in this thesis mainly estimated the radionuclide concentrations in the ecosystem components. However, the generated model results can also be used to estimate the exposure to biota, by conversion from radionuclide concentrations (Bq g C\(^{-1}\)) to energy per biomass (J kg wet weight\(^{-1}\)), and to humans, with dose conversion factors and assumptions of the consumption of contaminated food. External exposure may also be estimated from the results generated by ecosystem models since the concentrations in abiotic compartments are also assessed and the habitats of the organisms are well known.

Exposure to biota and humans consuming fish produced in the area around the final repository for operational waste after a hypothetical C-14 release were estimated from the modelling results obtained in Papers I and II (Kumblad 2001). The estimated exposures were very low both for biota (\(1 \times 10^{-12} – 1 \times 10^{-6}\) Gy) and for humans (0.6 µSv). These estimates were also similar to
those attained in a parallel modelling effort with BCF-based transfer models (Karlsson et al. 2001). The exposure to biota due to C-14 releases 2000 years from now would generally be higher compared to today ($1 \times 10^{-9} – 1 \times 10^{-6}$ Gy), although the exposure to fish, and consequently also the dose to humans, would be about the same (Kumblad 2001).

Aspects of model accuracy and reliability of the results

A real ecosystem or its development can never be fully described or predicted with a model without some degree of uncertainty. Nevertheless models are essential and useful tools in many disciplines. Model predictions are, for instance, often used as the basis for decisions that may be significant in terms of human, ecological or economic costs. It is therefore essential to evaluate the reliability of the model and to estimate the uncertainties of the predictions (Jørgensen 1994; Thiessen et al. 1999).

The accuracy of any model is a function of the parameter estimations, its structure and the mathematical algorithms used. The uncertainties of the model results may thus be divided into three categories: data, conceptual and representational uncertainties.

Most ecological data are collected for other purposes than for model development. It is also rare that areas are monitored to such extent that the collected data cover all parameters needed in ecosystem models. In the models, unknown fluxes are therefore often estimated from extrapolations from known parameters. These extrapolations introduce sources of uncertainty that may lead to false predictions, but may nevertheless be the only way to get estimations of likely magnitudes of unknown processes. The quality of the ecological input parameters used in the carbon flow model presented in Papers I (II and III) is high, as the majority of these data originate from field measurements in the area and this particular area has been thoroughly monitored. The probabilistic analysis performed in Paper III also shows that realistic variability of these parameters would have little effect on modelling results, probably due to the robust conceptualisation and ecosystem constraints. The variability in radionuclide concentration that may be caused by not accounted for interannual and seasonal variation in the carbon dynamics is,
however, smaller than the variability found in many transfer factor models. Uncertainties related to the radionuclide specific input parameters, i.e. BCF for plants and $K_d$, was shown to vary in importance for the different organism groups (Paper III).

The conceptualisation of the models presented in this thesis is robust. They were designed according to well known ecological principles and the structure of the ecosystem at the actual site. The food web in the carbon flow model is very simplified, but the ecological input parameters was compiled at a more detailed level (often at species level) and the resolution of the food web used for exposure modelling purposes is likely detailed enough.

The mathematical algorithms describing the metabolic processes in the ecosystem may be considered as being too simplistic. More sophisticated mathematical methods could enhance the resolution of the model results. The choice of mathematical techniques was however found to be of lesser importance for the applications of the models in this thesis because other parameters such as source term properties, biomass estimations or bioconcentration factors are likely to introduce larger uncertainties.

The degree of uncertainty of any model is evaluated in verification and validation processes. Model verifications are tests of the internal logic of a model and the performance of the algorithms used, i.e. that the calculations are accurate. Topics usually examined during verifications are whether the model responds logically to induced changes, its long-term stability, and mass conservatism (for mass balance models). The models in Papers I, II and III have all been successfully verified. The validation procedure tests how well the model output fits an independent set of data. Ideal validations can seldom be performed since there only rarely exist independent data sets for unique ecosystems. Predictions of future scenarios are of course also impossible to truly validate. However, attempts should always be made to validate models. The models described in this thesis have undergone four validation efforts of model results. The carbon flow model was validated against carbon dynamics described for an adjacent area (Paper I), and the CNP-dynamics (Paper II). The C-14 concentrations derived with the C-14 flow model were compared with measured C-14 concentrations outside Sellafield (U. K.) and doses calculated in a parallel exposure assessment with traditional transfer models (Karlsson et
The BCFs calculated with the generic radionuclide model were compared with BCFs from IAEA’s database (Paper III).

**Data requirements – the need for process oriented data**

The models in Papers I, II and III illustrate both the need and the possibility to use ecosystem models in ecological risk assessment of radionuclides. However, a major drawback is that this type of models requires a substantial amount of ecological knowledge and site-specific process-oriented indata. This type of data is unfortunately rather scarce within the field of radioecology (Whicker et al. 1999). The transfer functions for uptake and excretion of contaminants/radionuclides, needed for dynamic modelling, cannot be described with only a concentration ratio coefficient such as BCFs. Instead, descriptions of the uptake and excretion rates in relation to the properties of the organism and the environment are needed. In Paper III, the problem of not knowing the assimilation efficiencies or excretion rates for the radionuclides assessed was solved by assuming that there was no discrimination of radionuclides (in the food) in the uptake process, and by relating the elimination of the radionuclide to the elimination of carbon and a regulating factor (e.g. excretion coefficient, $K_e$). The effect of the regulating factor on the concentrations in the respective organism groups was then examined in model analyses. Bioaccumulation kinetics can also be explored experimentally. The uptake rate can for instance be estimated by measuring the increase in organism concentration exposed over a period of time. Similarly, elimination rates can be estimated by measuring the decrease in concentration in organisms after the exposure source has ceased. Bioaccumulation kinetic parameters of contaminants can be used to calculate bioaccumulation factors (BAFs). BAFs calculated in this way are based on biological processes in contrast to empirically derived bioconcentration factors (BCF) that are organism to media concentration ratios. Within the field of ecotoxicology, methodologies based on bioaccumulation kinetics are often used for chemicals such as pesticides (e.g. DDT), organochlorines (e.g. PCBs and PBDEs), and metals (e.g. Hg and Cd) (Landrum et al. 1996; Luoma and Fisher 1997), and applied in ecological risk assessments (e.g. Suter et al. 2000).
Bioaccumulation kinetics of radionuclides in benthic invertebrates

In Paper IV, the bioaccumulation kinetics and assimilation efficiencies of three sediment-associated radionuclides (Cr-51, Ni-63 and C-14) in three key benthic invertebrates were examined. There were two reasons for focusing on benthic organisms. First, potential radionuclide releases from the SFR-repository, which is located under the seabed, will likely enter the ecosystem via the sediment and benthic organisms will therefore have an important role in determining their fate. Moreover, coastal sediments are often highly contaminated with particle-reactive trace elements (e.g. Livett 1988; Luoma 1989; Borg and Jonsson 1996), which may be remobilised into aquatic food chains by processes such as bioaccumulation into benthic organisms (Fisher and Reinfelder 1995). Contaminant assimilation at the base of the food web can, in fact, be a major determinant of contaminant exposure and risks of toxic effects at higher trophic levels (Woodward et al. 1994; Munger and Hare 1997). Thus, understanding how and to what degree deposit-feeding benthic invertebrates bioaccumulate sediment-associated metals and radionuclides is important for both ecological risk assessment and management decisions in coastal ecosystems (Luoma and Fisher 1997; Chapman et al. 1998; Schlekat et al. 2002).

The selected organisms, the clam *Macoma baltica*, the amphipod crustacean *Monoporeia affinis*, and the priapulid worm *Halicryptus spinulosus*, are all key benthic organisms in the Baltic Sea. They differ substantially in terms of feeding ecology, feeding physiology, and digestive strategies, which often are important mechanisms behind observed differences in metal bioaccumulation among species (Lopez and Levinton 1987; Decho and Luoma 1991; Lee II 1991; Mayer et al. 2001). Very little is known of the bioaccumulation kinetics of metals into these species, as there have been few experimental studies (Sundelin and Eriksson 2001; Wiklund and Sundelin 2002).

The experiment presented in Paper IV was a two-part uptake and elimination study: (i) radionuclide uptake over 7 or 17 days by animals exposed to radio-labelled sediment, (ii) radionuclide elimination over 14 or 32 days by animals previously exposed to radio-labelled sediment, and (iii) determination of the assimilation efficiencies of the radionuclides (Figure 7). There were two different radionuclide treatments in the experiment: Ni-63 with Cr-51, and Ni-63 with C-14 incorporated in an algae detritus mixture.
Figure 7. Idealised uptake-elimination curve (radionuclide concentration in animals as a function of time), and experimental design for uptake and elimination phase for the two radionuclide treatments (Cr-51 + Ni-63 and C-14 + Ni-63) (Paper IV).

The bioaccumulation kinetic parameters were calculated according to methodologies often used (Landrum et al. 1992). The results from Paper IV support many of the assumptions used in the modelling Papers (I, II and III). For instance, since all radionuclides were found to be accumulated, benthic invertebrates are suggested to be important vectors for the transfer of sediment-associated radionuclides to higher trophic levels (e.g. fish and humans) via predation, and thus important to consider in exposure models. The radionuclides were accumulated to various extents, which imply that radionuclide specific parameters of, for example, assimilation efficiencies are needed in assessments. As the radionuclide uptake rates were found to be affected by changes in food availability, exposure models should be based on metabolic rates (kinetics) in combination with a whole ecosystem approach where both the demand and availability of food (carbon and nutrients) is assessed. Finally, the degree of accumulation for each radionuclide was similar between the three species, which suggests that benthic invertebrates having similar kinetics and position in the food web may be analysed together as a group rather than at the species level for the purposes of exposure modelling and ecological risk assessments.
Remobilisation of sediment-associated radionuclides by benthic invertebrates

Sediment-associated contaminants may not only be remobilised by bioaccumulation of radionuclides by benthic invertebrates and subsequent transport in the food web (Fileman et al. 1991), but also by sediment resuspension and bioturbation (Matisoff 1995; Peterson et al. 1996). Neither of these two processes was included in the models (Papers I, II or III), but the potential modelling errors due to omission of the bioturbation process were explored in Paper V. This was done by recreating a reoxygenation-recolonisation scenario in the laboratory, using anoxic laminated sediment cores collected in the field and the same three key benthic species as in Paper IV. The effect of this process on Cs-137 depth profiles was analysed. Fluorescent tracer was also added to the sediment surface of the cores to trace burial and downward transport of contaminants deposited on the sediment surface. The bioturbation experiment ran for 25 days.

The laminations were still visible at the end of the experiment, though the sediment had clearly been disrupted by the activities of the animals, particularly by *H. spinulosus*. Although there were visual differences between the sediment effects of the three species, no significant differences between the treatments could be found for either Cs-137 or fluorescent tracer. The fluorescent tracer was also transported downwards in the Azoic control. Possible explanations for this lie with biogeochemical processes occurring after reoxygenation or with anoxia tolerant meiofauna that could have been present in the anoxic cores.

The sedimentary history of the area, deduced from the Cs-137 profiles and Pb-210 dating, showed that it was likely that the area had been subject to resuspension and redeposition of sediment by physical processes. This made it impossible to use the Cs-137 profiles as a tracer of bioturbation during the experiment, but also suggested that, at this site, physical processes are likely to be far more important than biological processes in the redistribution of contaminants on a decadal timescale. Thus, the results in Paper V suggest that the errors introduced in Papers I, II and III by omitting bioturbation effects are negligible.
Concluding discussion

In this thesis a methodology based on ecosystem modelling was used to estimate the transport and fate of radionuclides in the coastal environment above the underground nuclear waste repository (SFR) in Öregrundsgrepen, Baltic Sea. The model results were similar to those retrieved in a parallel modelling effort with BCF-based transfer models and in the same range as empirically derived data for similar organisms for most radionuclides (IAEA 1985; Karlsson et al. 2001; Paper I and III). This demonstrates that the ecosystem modelling method gives sufficient estimates of the environmental transfer of radionuclides and is at least as reliable as estimates generated by BCF-based models, which normally are used for these purposes. The ecosystem models not only produced results similar to BCF models, but could also provide results for variables and explore possible scenarios that non-mechanistic models are unable to assess. For instance, the methodology allowed estimations of radionuclide exposure to all biota groups in the system, and not only to those organisms for which BCF values were available. It was also possible to analyse how the ecosystem dynamics influenced the radionuclide fate. These analyses included estimations of the time needed to reach steady-state conditions in the receiving components and the ecological half-life after the simulated radionuclide release had ceased (e.g. Paper I). The influence of environmental processes such as water exchange rate on the exposure of the organisms in the ecosystem could also be explored (Paper I and III), as well as how the differences in radionuclide specific mechanisms affected the exposure of biota (Paper III).

Due to the mechanistic structure of the models and the fact that they are based on intrinsic ecological processes and constraints of the ecosystem, they could also be scaled for predicted future changes in environmental properties in a realistic way (e.g. changes in bathymetry and habitat, Paper II). In time frames of several thousand years, changes in parameters such as temperature, salinity, nutrient load and introduction of new species may influence the development of the ecosystem. Increased temperature may enhance the metabolic turnover in the system and thus the uptake of radionuclides. Changes in temperature might also influence the ecosystem structure. Salinity changes would likely shift the ecosystem structure to be similar to areas located just north or south of the modelled bay. These types of environmental changes may be handled in
ecosystem models. The effects of the introduction of alien species or human interactions with the environment are on the other hand more difficult to predict. However, since ecosystem models are based on the constraints of the system itself, possible ranges of consequences can be identified.

Although the Papers in this thesis have illustrated that there are many advantages in using an ecosystem approach, there are also some drawbacks that need to be mentioned. In Table 2 the findings in this thesis concerning the use of ecosystem models compared to BCF-based exposure models are summarised.

Table 2. Some characteristics of transfer factor based models and ecosystem models

<table>
<thead>
<tr>
<th>Ecosystem models</th>
<th>BCF-based transfer models</th>
</tr>
</thead>
<tbody>
<tr>
<td>Complex to use</td>
<td>Easy to use</td>
</tr>
<tr>
<td>Dynamic, mechanistic and mass balanced</td>
<td>Steady-state and not mass balanced</td>
</tr>
<tr>
<td>Require a considerable amount of site-specific ecological parameters, but these are generic for all radionuclides</td>
<td>Require few or no site-specific parameters</td>
</tr>
<tr>
<td>Require only a few radionuclide specific factors</td>
<td>Require a multitude of radionuclide specific factors (of which many are uncertain)</td>
</tr>
<tr>
<td>Model everything in the ecosystem, e.g. concentrations in organism and abiotic media, flows, distributions</td>
<td>Only estimate the concentrations in organisms for which there are available BCFs (mainly for organisms of interest for human consumption)</td>
</tr>
</tbody>
</table>

A major advantage of the use of mass balanced models in exposure assessments is that as long as the carbon flow model is adequate, unrealistic results cannot be produced. For instance, in the models presented in this thesis, the radionuclide uptake was constrained by the metabolic uptake rates of the respective organism group, i.e. primary production or consumption, and the radionuclides available for uptake in the respective compartment, i.e. in DIM or the food sources. As a consequence, biota may never accumulate more radionuclides than the total amount released into the system, or to a larger extent than the total amount of matter ingested (food). This may be self-evident, but is not always the case for BCF-based transfer models, as these are neither mass balanced nor consider recirculation processes in the system or
changes in radionuclide availability. This problem with BCF-based models was also identified in a validation project of radioecological transfer models (IAEA 1996), where the modelling results varied by up to five orders of magnitude for the same type of simulations.

Ecologically sound models not only require ecological knowledge, but also a large amount of site-specific ecosystem data, which may be difficult and costly to attain. This may be a major drawback for the possibilities of using this approach. However, safety assessments for nuclear facilities such as waste repositories will eventually always be site-specific, and a well-documented description of the ecosystem is likely to be required before building permits can be given. Many parameters required for ecosystem modelling can also be estimated and extrapolated from abundance data on species present.

Another fundamental problem in safety assessments of radionuclides is that radionuclides may differ significantly in chemical and physical properties. Their fate in the environment can therefore not strictly be linked to the flow of, e.g., carbon, since processes such as absorption, excretion and adsorption are different for them compared to carbon (except for C-14). A carbon flow model can nevertheless provide a frame for possible uptake and transfer pathways of radionuclides and limit the modelling to realistic ranges of the uptake (as discussed above). One method to handle this problem was explored and discussed in Paper III, which may exemplify a step towards a new methodology that can be used in ecological risk assessments of radionuclides.

The overall purposes of the modelling studies presented in this thesis were to examine the possibility of applying an ecosystem approach in radionuclide assessments, and to evaluate if the approach overcomes the problems with BCF-based steady state models. The presented Papers show both that this approach was possible to use the ecosystem approach to assess the exposure to biota, and that it can handle many of the identified modelling problems. Consequently, the findings in this thesis suggest that both national and international assessment frameworks for protection of the environment from ionising radiation would benefit from striving to adopt methodologies based on ecologically sound principles and modelling techniques.
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