Journal of Environmental Radioactivity 169-170 (2017) 85–97

Contents lists available at ScienceDirect

Journal of Environmental Radioactivity

journal homepage: www.elsevier.com/locate/jenvrad

Application of an ecosystem model to evaluate the importance of different processes and food web structure for transfer of 13 elements in a shallow lake

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A R T I C L E I N F O

Article history:
Received 25 May 2016
Received in revised form
25 December 2016
Accepted 29 December 2016

Keywords:
Concentration ratio
Bioaccumulation
Model
Lake
Risk assessment
Food web

A B S T R A C T

In environmental risk assessments of nuclear waste, there is need to estimate the potential risks of a large number of radionuclides over a long time period during which the environment is likely to change. Usually concentration ratios (CRs) are used to calculate the activity concentrations in organisms. However, CRs are not available for all radionuclides and they are not easily scalable to the varying environment. Here, an ecosystem transport model of elements, which estimates concentrations in organisms using carbon flows and food transfer instead of CR is presented. It is a stochastic compartment model developed for Lake Eckarfjärden at Forsmark in Sweden. The model was based on available data on carbon circulation, physical and biological processes from the site and identifies 11 functional groups of organisms. The ecosystem model was used to estimate the environmental transfer of 13 elements (Al, Ca, Cd, Cl, Cs, I, Ni, Nb, Pb, Se, Sr, Th, U) to various aquatic organisms, using element-specific distribution coefficients for suspended particles ($K_{d\ PM}$) and upper sediment ($K_{d\ sed}$), and subsequent transfer in the foodweb. The modelled CRs for different organism groups were compared with measured CRs from the lake and literature data, and showed good agreement for many elements and organisms, particularly for lower trophic levels. The model is, therefore, proposed as an alternative to measured CR, though it is suggested to further explore active uptake, assimilation and elimination processes to get better correspondence for some of the elements. The benthic organisms (i.e. bacteria, microphytobenthos and macroalgae) were identified as more important than pelagic organisms for transfer of elements to top predators. The element transfer model revealed that most of the radionuclides were channelled through the microbial loop, despite the fact that macroalgae dominated the carbon fluxes in this lake. Thus, element-specific adsorption of elements to the surface of aquatic species, that may be food sources for organisms at higher trophic levels, needs to be considered in combination with generic processes described by carbon fluxes.

1. Introduction

In environmental risk assessment of intermediate or high level radioactive waste, several challenges arise. The assessment considers a hypothetical release of more than 40 different radionuclides into the environment several millennia into the future (Kautsky et al., 2013). To estimate the activity concentration in aquatic organisms, for instance fish, a concentration ratio (CR) is classically used which is based on measured concentrations in fish and surrounding water. However, for many radionuclides there are either no available CRs derived from measurements or those that exist are from, for example, nuclear weapon test sites where the ecosystems or environments are different from the site of interest, and data may be scarce and sometime of little relevance for the investigated site (IAEA et al., 2009). Another approach is to estimate CRs from stable element concentrations at the site of interest (Bradshaw et al., 2012; Konovalenko et al., 2014; Tröjbom et al., 2013), which gives current realistic values for a spectrum of elements in natural ecosystems. Radionuclides are assumed to have

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http://dx.doi.org/10.1016/j.jenvrad.2016.12.016

the same chemical properties as their stable isotopes, except for the slight differences resulting from the difference in mass (Santschi and Honeyman, 1989). In the aquatic environment, radionuclides are therefore transported and taken up by biota in a similar way to their stable element analogues (Sazykina, 2000). Stable element data are commonly used in assessment of radionuclide transfer to organisms (Brown et al., 2016; Copplestone et al., 2013). Simple CR-based approaches can however be difficult to extrapolate to future conditions with a different climate or different sized ecosystems (Lindborg et al., 2013), since CRs combine element-specific processes (e.g. sorption, uptake) with element-independent processes, such as food consumption and site-specific water turnover. Thus, it is advantageous to use a method which can be scaled to ecosystem properties, and where element-specific processes are separated from generic processes which are equally valid for all elements. Several successful attempts have been made for coastal ecosystems to show that element uptake can be estimated using ecosystem models and a limited number of measurable element-specific parameters (Kumblad et al., 2006; Sandberg et al., 2007; Konovalenko et al., 2014), but they have not been established for lakes where the microbial loop and turnover of dissolved organic carbon (DOC) are usually important (e.g. Tranvik, 1992; Andersson and Kumblad, 2006).

In ecotoxicology and radioecology, there is also increasing recognition that ecosystem models for radionuclide or element transport and cycling should be based on mathematical equations describing processes, rather than solely on empirical parameters, such as transfer factors (Hinton et al., 2013). Process-based models are more robust, since they take into account uncertainty and environmental variability (Cuddington et al., 2013), and allow the identification of processes that are of most importance to radionuclide transfer (Belharet et al., 2016; Kirchner and Steiner, 2008). Transfer models for radionuclides, metals or other elements are essentially based on similar principles. Radionuclide and heavy metal transport is quite well described using modelling for simple aquatic food chains (e.g., phytoplankton – zooplankton – prey fish – predatory fish) (Sazykina, 2000; Kryshev et al., 2003; Heling and Bezhenar, 2009), but relatively few studies have been conducted on the cycling and transport of radionuclides in more complex food webs, for example including microbes, aquatic plants, invertebrate grazers and benthic-pelagic interactions. Often, the only primary producer considered is phytoplankton, but benthic plants and microalgae may comprise a major part of the biomass of shallow lake ecosystems. In addition, there can be significant accumulation of some elements or isotopes (e.g. Tc-99, Ca, etc.) in some species of aquatic plants and algae (Fukuda et al., 2014), which in turn may be consumed by grazers (e.g. crustaceans, molluscs), and ultimately, fish. Studies of uptake some metals (e.g., Ag, Am, Co, Cd, Cu, Se, Tc, Hg) by aquatic organisms suggest that dietary metal exposure can be an important source of accumulation (Wang and Fisher, 1998; Xu et al., 2001; Griscom et al., 2002; Xu and Wang, 2004). Thus, bioaccumulation of contaminants may be underestimated if these compartments of the ecosystem are not included in the model. Another pathway of exposure of aquatic benthic organisms (e.g. filter feeding, soft-bottom macrofauna) may be via sediment, benthic microbes and suspended particles, but this pathway is seldom incorporated in element transport models.

This is in contrast to ecological/ecosystem modelling, where many models of different levels of complexity describing carbon circulation have been developed (Jorgensen et al., 1978; Jorgensen et al., 1981) including, for example, physical-microbial food web models (Gall et al., 2009). However, such models have only addressed carbon and energy fluxes without consideration of other elements or contaminants in the system. For lakes, most radionuclide or element transport models have examined the behaviour of a few or single elements. These are usually toxic and/or potentially biomagnifying metals such as Hg and Cd (Cardwell et al., 2013) or, in the case of radionuclides, the dominant fallout biomagnifying isotope Cs-137 and/or bioaccumulated isotope Sr-90 (Zheleznyak et al., 1997; IAEA, 2000; Smith et al., 2003).

The primary aim of this study is the development of element transport model for a lake that is built on a process-based ecosystem model of carbon flow and capable of estimating the transport of multiple elements in a complex food web. The purpose of the model development was to investigate the possibility of modelling the transfer and turnover of 13 elements for eleven functional groups of lake organisms and to calculate CRs for the functional groups, a capability that would be useful for estimations of the fate of future theoretical radionuclide releases from a nuclear waste disposal facility. A secondary aim was to identify which biota compartments and processes were most important in element cycling, and especially in the transfer and accumulation of elements in biota.

2. Methods

2.1. Site description

Lake Eckarfjärden is a small, shallow, oligotrophic, hard water lake located 2 km south of Forsmark (60°22′ N, 18°12′ E), near the Baltic Sea coast in the province of Uppland, Sweden (Fig. 1). The lake is of interest from a radionuclide transfer perspective as it lies close to the Forsmark Nuclear Power Plant (NPP) and the site for a proposed deep repository for high level radioactive waste (Lindborg, 2010). It has, therefore, been the subject of detailed ecological investigations by scientists associated with the Swedish Nuclear Fuel and Waste Management Co. (SKB) (e.g., Brunberg et al., 2002; Andersson, 2005; Andersson and Sobek, 2006). The submerged macroalga, *Chara* sp., covers approximately 50% of the bottom area of L. Eckarfjärden (Andersson and Kumblad, 2006). The lake is also characterized by a thick benthic microbial mat (1–10 cm deep) consisting of microphytobenthos and heterotrophic bacteria (Andersson, 2010). The biomasses of the major groups are found in Table 1. The detailed hydrochemistry of L. Eckarfjärden is given by Andersson (2010) and Andersson and Brunberg (2006), but the main chemical characteristics are as follows: the lake water pH is 8 ± 0.7; water colour is moderate (mean absorbance at 420 nm 0.147 ± 0.043 cm^{-1}); alkalinity is 2.5 ± 0.4 meq L^{-1}; the water concentrations of Ca and K are 44 ± 10 mg L^{-1} and 2 ± 0.3 mg L^{-1}, respectively; and the water temperatures ranges between 0.5 and 20 °C.

2.2. Modelling

The model developed in this study assumes that elements follow flows of organic matter in the food web, proportionally to carbon flows. Additional processes include adsorption of elements from water to particles and to the surfaces of organisms. The underlying carbon model is process-based and builds on earlier carbon budgets by Andersson and Kumblad (2006), Andersson and Sobek (2006) and Andersson (2010). The model was partly descriptive, rather than predictive, because site-specific field data were not always available as input data for modelling or for model validation. The key concepts and general processes of the model are identical to an earlier radionuclide transport model for a coastal area (Kumblad et al., 2003; 2006; Konovalenko et al., 2014), though in this paper, additional processes and compartments were considered that are characteristic for the lake. The ecosystem of Lake Eckarfjärden was subdivided into 16 compartments (Fig. 2, Table 1). Two compartment models are described below: Carbon